SURFACE MODIFICATIONS OF REVERSE OSMOSIS MEMBRANES FOR REMOVAL OF BROMIDE AND REDUCTION OF FOULING

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ABSTRACT

Surface Modifications of Reverse Osmosis Membranes for Removal of Bromide and Reduction of Fouling

Joseph Seo

Reverse Osmosis (RO) is widely used for water reuse and desalination applications. Although RO membranes are known for their high salt rejection and practical permeate flux, their performance can be impaired by fouling and their removal for some disinfection byproducts and their precursors is not sufficient to meet the standards. RO membrane modifications have been widely studied to overcome these limitations. In this research, RO membranes were grafted with cationic polymers to induce a positive charge on the RO membrane surface. This medication aimed at enhancing the rejection level of the negatively charged bromide ions, which are precursors for the formation of disinfection byproducts. The results showed that the modified (positively charged) RO membranes achieved lower rejection for bromide ions compared to the unmodified ones. The rejection efficiency dropped from 94.5% to 82.0% after modification. This behavior was likely a result of increased concentration polarization of the bromide ions at the membrane surface due to the increase in attraction energy of the modified membrane. Calculations based on the film theory showed that the concentration of bromide ions at the surface of the modified membrane was 1370.6 ppm compared to 1306.6 ppm at the surface of the unmodified membrane. This further supports that assumption that the positive charge on the membrane surface has attracted the bromide ions from the bulk solution. However, this attraction energy was not sufficient to keep the bromide ions attached to the membrane surface and prevent their diffusion across the membrane as originally hypothesized in this research investigation.

Although the goal of the modification in the current study (i.e., enhancing removal of bromide ions) was not met, the permeate flux of the modified membrane was improved compared the unmodified one. The study findings highlight a critical aspect for consideration in future membrane modification research, which is the need for a more comprehensive performance evaluation of the modified membranes beyond the target goals. For example, many modifications to increase RO membrane flux production have been successful but there may have been unintended consequences of such modifications that compromise the efficiency of the membranes to reject certain pollutants. Therefore, it is recommended that future research on membrane modifications use a more comprehensive approach for evaluating the effects of such modifications beyond the target goals (e.g., flux improvement).

In addition to the experimental work conducted in this study, a multi-criteria decision analysis was performed to prioritize research on surface modifications of reverse osmosis membranes. It was found that surface modifications have been mainly focused on reducing membrane fouling and to a much lower extent on removal of disinfection byproducts and their precursors. The RO membrane modification alternatives for fouling reduction and N-Nitrosodimethylamine (NDMA) removal were ranked based on multiple criteria using the Analytical Hierarchy Process (AHP) and the Technique for Order Preference by Similarity to Ideal Solution (TOPSIS). This multi-criteria decision analysis process resulted in the identification of the top 5 promising modifications to reduce fouling and improve NDMA rejection. Grafting and coating the RO membranes with complex polymeric salts were the highest ranked modification approaches to reduce fouling. On the other, heat treatment of RO membranes achieved the highest NDMA rejection (98%) compared to 10 - 40% for traditional RO membranes. However, this technique

was the second highest ranked modification approach for NDMA removal because it scored lower for other evaluation criteria.

Keywords: Reverse Osmosis, Desalination, Portable Water Reuse, Membrane Modification, Bromide, NDMA, Surface Grafting, AHP, TOPSIS, Multi-criteria Decision

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1.1 Introduction

Fresh water resources are vital for sustaining life. As water pollution is growing globally, countless communities are losing access to drinkable water [1]. As a result, a growing emphasis exits on development of effective technologies for potable water reuse [2]. Indirect potable reuse involves sending reclaimed water to freshwater bodies in the environment before further treatment and distribution to the consumers. For direct potable reuse, highly purified wastewater is directly to the consumers. However, direct potable reuse facilities are limited in the United States and they are only allowed to operate under emergency situations [1]. In California, indirect potable reuse facilities use a combination of reverse osmosis (RO) and advanced oxidation processes to meet the requirements for groundwater injection [3]. Although California has allowed surface water augmentation through SBDDW-16-02 Surface Water Augmentation Regulations, direct potable water reuse is not permitted [1].

Reverse osmosis (RO) is one of the most reliable technologies for potable reuse applications. However, one of the limitations of reverse osmosis membranes is their inadequate removal removal efficiency for harmful disinfection byproducts and their precursors such as N-Nitrosamine (NDMA) and bromide [4]. Disinfection byproducts are potential carcinogens, mutagens, and toxicants [5, 22].

Bromide ion is a growing concern for water treatment and desalination processes as it is a precursor for the formation of disinfection byproducts [4, 5]. Bromide ion is found in various water sources including groundwater, seawater, and wastewater [5, 9, 10]. It is a naturally occurring constituent that can reach groundwater from seawater intrusion [9]. It can also be found in anthropogenic pollution from using organic bromyl compounds in agricultural applications [10]. Water treatment including potable reuse processes require disinfection of the treated water. Ozone is one of the effective alternative disinfectants used in water treatment. However, when bromide is present in the water to be disinfected, it oxidizes and forms bromate [6]. Because bromate is a carcinogen, it is crucial to remove bromide from the water before disinfection with ozone [7, 8]. It is desirable to remove bromide ions before disinfection because once bromide ion oxidizes and forms DBPs, their removal is uneconomical and difficult [11]. Furthermore, ozone is a commonly used oxidizer in advanced oxidation processes, which typically follow RO systems in potable reuse facilities [11]. If reverse osmosis membranes do not sufficiently remove bromide ions, high levels of bromate could form in the water during the advanced oxidation process.

During the RO process, water or wastewater is driven under pressure to pass through semi-permeable membrane and allow water molecules to pass through without the contaminants [12]. Concentration polarization takes place during RO processes as shown in Figure 1.1. As the contaminants accumulate on the membrane surface, the concentration becomes higher at the surface compared to that in the bulk feed (i.e., polarization occurs). This phenomenon reduces the water flux through the RO membrane and decreases the salt rejection (i.e., increase solute

transport across the membrane) [13, 21]. Therefore, concentration polarization impacts the rejection of bromide by RO membranes.

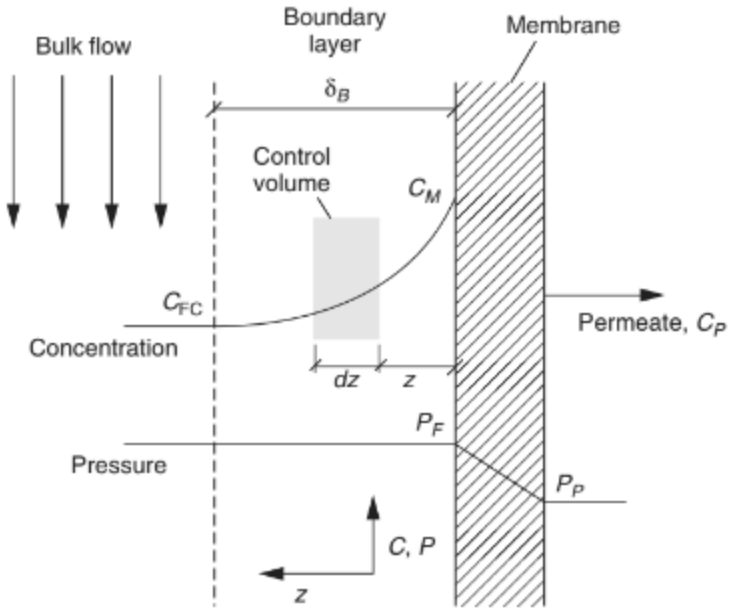


Figure 1. 1 Schematic of the concentration polarization phenomenon [22]. The curve created by C_{FC} and C_M represents the increasing concentration of contaminant.

There is limited research on improving bromide ion rejection by RO membranes. A study conducted by Watson et al. reported rejection of bromide ranging from 93 to 99.3% [23]. Although the membranes achieved high rejection, the operating pressure required to achieve 99.6% bromide ion rejection was ~940 psi. Constantly pumping large volumes of polluted water at high pressure is mechanically and economically inefficient [23]. Depending on membrane type used (e.g., seawater RO (SWRO) membranes) bromide ion concentration in the permeate may reach 1000 μg/L, which is 100 times higher than the 10 μg/L guideline set by WHO [14]. Because seawater RO membranes have lower removal efficiency for bromide, SWRO desalination plants in Australia generally use two-stage RO systems [14].

The objective of this study was to investigate the effect of modifying the surface charge of reverse osmosis membranes on the removal efficiency of bromide ion. Polyamide RO membranes, widely used in large-scale RO systems, are negatively charged due to the deprotonation of carboxyl groups on the membrane surface [15]. This implies that bromide ions, which are negatively charged ions, should be rejected by the membrane surface. However, with these repulsion forces in place, RO membranes do not remove bromide ions to sufficient levels to avoid the formation of DBPs in subsequent ozonation processes. In this study, reverse osmosis membranes were grafted with branched polyethylenimine (PEI) to make the membrane surface positively charged. PEI is a polymer with amine groups that protonate in solution and exhibit a positive charge of up to 23.3 meq/g in aqueous solution depending on the water pH [16]. By modifying the RO membrane surface charge to positive, it is hypothesized that negatively charged bromide ions will strongly bind to the positively charged RO membranes with an electrostatic force that is sufficient to limit the transport of bromide across the RO membrane.

1.2 Materials and Methods

1.2.1 Materials

Flat sheet reverse osmosis membrane coupons, 140 mm x 190 mm (5.51" x 7.48"), was purchased from Sterlitech Corporation (WA, USA). The membrane is a SW30HR model manufactured by Dow Filmtech™. This is a polyamide thin film composite (TFC) membrane designed for seawater feed with high rejection of up to 99.6% and can operate in a pH range of 2-11 at 25°C**.** The membrane is rated to produce a flux of 17-24 gfd at 800 psi. N-(3-

Dimemthylaminopropyl)-N-ethylcarbodiimide hydrochloride (EDC-HCl) 98% was purchased from Sigma-Aldrich. N-hydroxysuccinimide (NHS) 98% was purchased from Acros Organics. Branched PEI (MW 1200) was purchased from Polysciences, Inc.

1.2.2 Membrane Modification

The membrane surface was modified following the method reported by Xu et al. [15]. Briefly, a membrane coupon was placed in a beaker with 120 ml of DI water mixed with 0.138 g of EDC-HCl. After 10 minutes, 0.066 g of NHS was added to the beaker containing the RO membrane and EDC-HCL solution. This mixture was kept equilibrating for another 15 minutes and then 6 g of branched PEI (molecular weight 1200) was added. The RO membrane was left in this chemical mixture in a sealed zip-lock bag in a dark environment at room temperature for 12 hours. The membrane was then removed from the mixture, washed, and stored in DI water in a zip-lock bag until use.

EDC-HCl is used in this grafting process as an activating agent and NHS as a stabilizer to allow amine bonds of branched PEI to react with carboxyl groups on the surface of RO membranes [15]. Theoretically, when the carboxyl groups on the membrane surface and EDC-HCl react, o-acylisourea ester intermediate is formed, which is highly reactive and allows bonds to form (Figure 1.2). Then, the addition of NHS results in the formation of a semi-stable NHS-ester intermediate, which makes the carboxyl group unstable. This intermediate serves as the reacting bridge to connect the amine groups of branched PEI to the membrane surface.

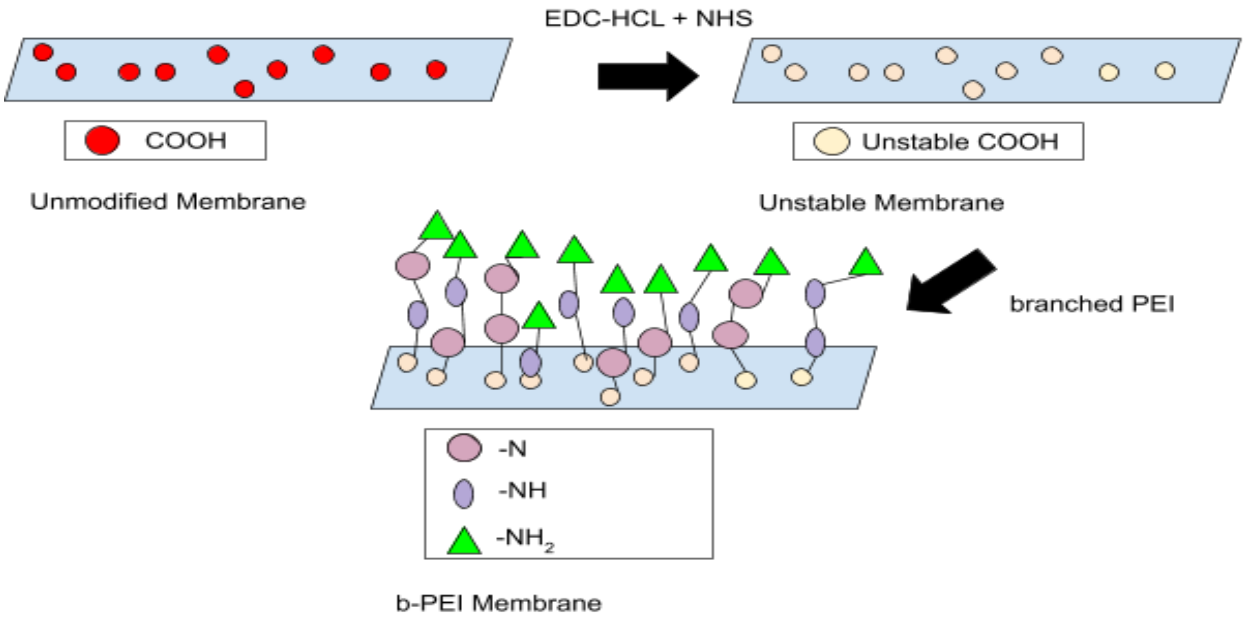


Figure 1. 2 Schematic of the PEI grafting process to alter the RO membrane surface chemistry.

1.2.3 Surface Characterization of the Modified Membrane

Fourier transform infrared spectroscopy (FTIR) was used to verify that the membrane surface was coated with PEI. The FTIR measurements were performed using a Jasco FTIR-4600 instrument (MD, USA). The resolution was set to 4 cm^{-1} and the wavenumber ranged from 400 to 4000 cm^{-1}. When a surface is exposed to infrared light, the covalent bonds of the molecules start to resonate, and the resonance wavelength can be used to detect the presence of functional groups and element pairings on the sample surface [24].

1.2.4 Reverse Osmosis Testing Apparatus

A Sepa cell system manufactured by Sterlitech (WA, US) was used for testing the efficiency of the modified reverse osmosis membranes for removing bromide ions. The Sepa Cell is a 316 stainless steel bench-scale membrane cell that allows membranes to filter water in cross/tangential flow mode under a range of operating pressures. It has a membrane active area of 140 cm^2 and maximum operating pressure of 1000 psi. A hydraulic pump is attached to pressurize the cell and prevent water from leaking.

A schematic of the reverse osmosis test apparatus is presented in Figure 1.3. The membrane coupons were placed inside the membrane cell. The membrane was sandwiched in between a feed spacer and a permeate carrier to mimic the conditions used in spiral wound RO membrane elements. The feed water was pumped into the cell in a cross flow mode. The concentrate was re-collected into the same feed container while the treated water (permeate) was collected in a separate beaker for sampling. The RO system was operated at 300 psi.

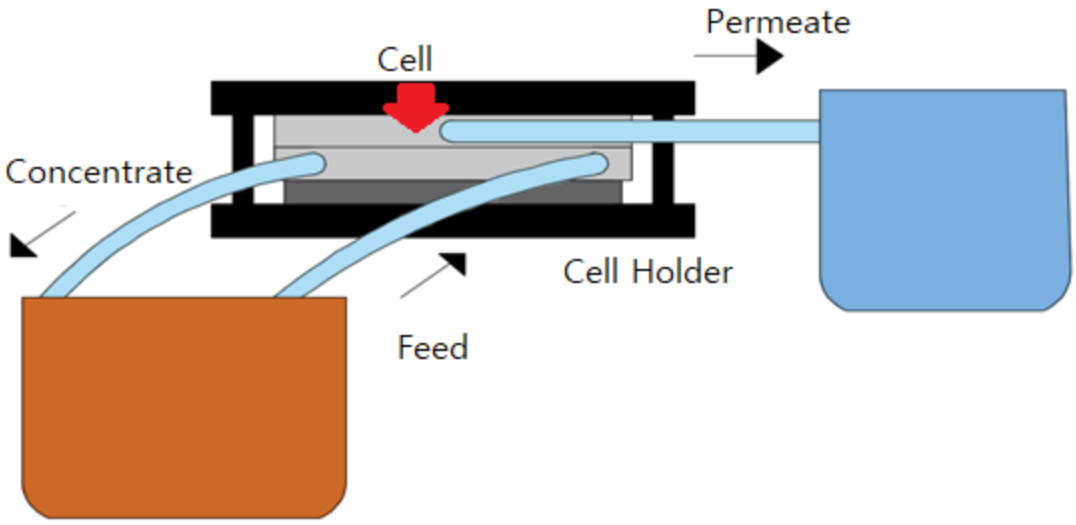


Figure 1. 3 Schematic of the water flow path in the Sepa cell system.

The membrane cell was flushed with DI water before and after each test. The feed water contained 1000 ppm bromide in DI. The feed solution was pumped into the cell system and samples of the permeate were collected over time. The first 50 ml of permeate collected were discarded to avoid any residual effluent from previous runs. Control experiments were also conducted using unmodified membranes to enable comparison of the effect of RO membrane modification on removal of bromides. All experiments were conducted in triplicates.

1.2.5 Permeate Measurements

The concentration of bromide ions in the permeate samples was tested using ion chromatography to determine the rejection efficiency of modified and unmodified RO membranes. The ion chromatograph (IC), Dionex DX120, was calibrated using standard solutions of sodium bromide with Br^- concentrations ranging from 10 – 100 ppm. Aliquots of 5 mL permeate samples were injected into the IC column and the area under the ion chromatogram curve was used for

calculating the concentrations. The total dissolved solids (TDS) and temperature of the permeate were measured using a PC400s Portable pH/Conductivity Meter kit (Apera Instruments). The permeate flux was determined by measuring the permeate volume collected into a beaker for a period of 8 minutes. The flux was calculated using Equation 1.

$$J_w = \Delta V / A \Delta t \qquad (1)$$

Where, J_w is the water flux in the unit of L m^{-2} h^{-1} and $\Delta V/\Delta t$ is the change in volume over a given period.

1.2.6 Concentration Polarization Calculations

Concentration polarization is the increase of solute concentration near the surface of the membrane compared to that in the bulk feed. The concentration of bromide at the surface of the membrane was predicted using Equation 2, which is a mathematical relationship that describes the concentration polarization phenomenon [21].

$$\frac{C_M - C_P}{C_{FC} - C_P} = e^{Jw/k_{Cp}} \qquad (2)$$

Where, C_M is the concentration of solute at the membrane surface, C_P is the concentration of solute in the permeate, C_{FC} is the concentration of solute in the bulk feed water, J_W is the water flux, and k_{CP} is the concentration polarization mass transfer coefficient. These parameters were

determined experimentally except k_{CP}, which was calculated using correlations of dimensionless parameters using Equations 3 – 5.

$$k_{CP} = 0.023 \frac{D_L}{d_H} (Re)^{0.875} (Sc)^{0.25} \quad (3)$$

$$\text{Re} = \frac{\rho v d_H}{\mu} \quad (4)$$

$$\text{Sc} = \frac{\mu}{\rho D_L} \quad (5)$$

Where, Re is Reynolds number, Sc is Schmidt number, v is velocity of water in the feed channel, ρ is the water density, μ is the dynamic viscosity of water and d_H is the hydraulic diameter. The units are in metric system.

Using the above equations, the concentration of bromide at the surfaces (C_M) of modified and unmodified membranes was predicted. The research hypothesis can be tested by comparing C_M for the modified and unmodified membranes. The positively charged (modified) membranes should have higher concentration of negatively charged bromide ions at the membrane surface due to the electrostatic attraction compared to negatively charged unmodified membranes if the research hypothesis is true. It is noted that the remaining part of the hypothesis that the charge interactions are strong enough to prevent bromide from crossing the membrane surface was tested by measuring the bromide concentration in the permeate.

1.3 Results and Discussions

1.3.1 Membrane Characterization

The modified membrane was tested using FTIR to confirm that PEI was successfully grafted on the RO membrane surface. Figure 1.4 shows the transmittance peaks of the FTIR spectrum of the modified membranes. The spectrum shows N-H stretching band around 3250 cm^{-1} (the first red circle from left) and the amine group peak around 1650 cm^{-1} (second red circle). Both bands are indicative of the presence of amine groups. Other bands that are relevant to PEI were C-H stretching around 1500 cm^{-1} and C-N bending band around 1350 and 1100 cm^{-1} (third red circle). Among the aforementioned indicators, the band of the amine group is the strongest indicator that the membrane was functionalized with PEI and thus, is expected to exhibit positive surface charge when it is in contact with water.

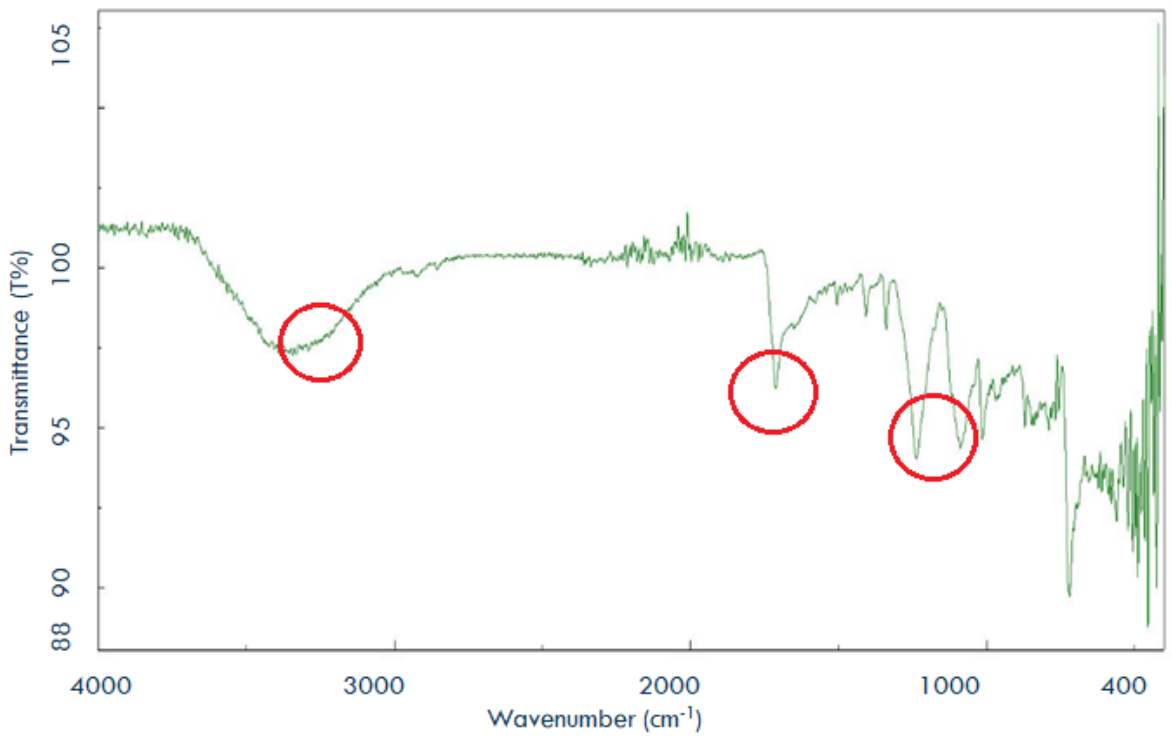
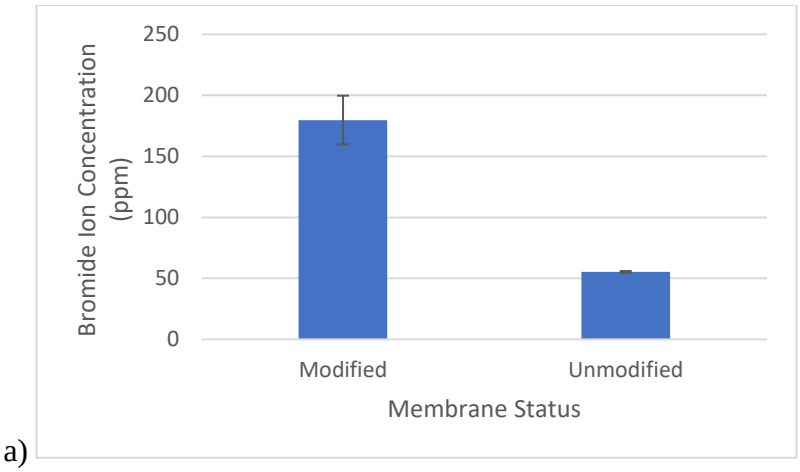


Figure 1. 4 The FTIR spectrum of the RO membrane grafted with PEI.

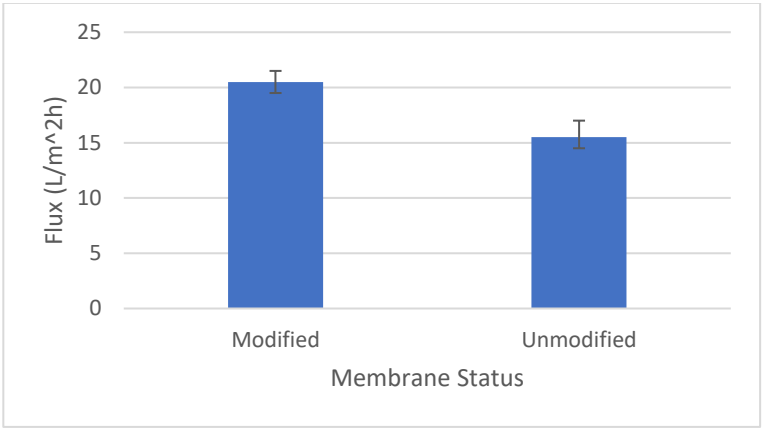
1.3.2 RO Membrane Performance

Bromide ion concentration in the feed was 1000 ppm. The bromide ion concentrations in the permeate and the water flux of the modified and unmodified membranes are presented in Figure 1.5 (a and b). The modified membrane permeates had an average Br^- concentration of 179.7 ppm compared to 55.2 ppm for the unmodified membrane. This shows 82.0% Br^- removal for the modified membrane compared to 94.5% for the unmodified membrane. On the other hand, the modified membrane had a higher water flux (20.5 L/m^{2}h) compared to that of unmodified membrane (15.5 L/m^{2}h). The increase in flux after membrane modification with PEI was also observed in previous studies [15]. The researchers hypothesized that the PEI increased

hydrophilicity of the membrane surface, allowing the diffusion of water molecules to happen more readily.



a)



b)

Figure 1. 5 Comparison of the membrane modification performance a) bromide ion concentration in the permeate and b) permeate flux.

The concentration of bromide ions at the membrane surface was predicted using Equations 2 to 5. These calculations were performed to test the theory that the attraction forces between the positively charged membrane and the negatively charged bromide ions would lead to accumulation of higher concentration of bromide ions at the boundary of the modified membrane compared to the unmodified one. When experimental data were not available, assumptions were

made to perform the calculations using Equations 2 to 5. For calculating Reynold's number, the density and dynamic viscosity of water at 69 °F were 998 kg/m^3 and 1.0 x 10^{-3} kg/m.s, respectively. The hydraulic diameter and water velocity were assumed 1.72 mm and 0.15 m/s, respectively [21]. Diffusivity of bromide ions in water at 69 °F was 1.6 x 10^{-4} m^2/day.

The experimentally determined C_P for the modified membrane was 179.7 ppm and it was 55.2 ppm for the unmodified membrane. C_{FC} was 1000 ppm for both modified and unmodified membranes. The measured water flux values were 20.5 L/m^2h and 15.5 L/m^2h for the modified and unmodified membrane, respectively. Using the aforementioned values, the calculations resulted in a C_M of 1370.6 ppm for the modified membrane compared to 1306.6 ppm for unmodified membrane. These results show that the concentration of bromide ions was higher at the surface of the modified membrane due to the attraction forces between the PEI molecules and the bromide ions.

The results obtained in this study likely indicate that the PEI grafted on the surface of the membrane was effective in attracting the bromide ions to the membrane boundary layer, as was hypothesized. However, the magnitude of electrostatic forces was likely not sufficient to prevent diffusion of the bromide ions across the RO membrane leading to a higher concentration in the permeate for the PEI modified membranes. Further research is needed to improve the efficiency of removal of bromides using RO membranes.

1.4 Conclusions and Future Recommendations

The membrane characterization preformed as well as the RO experimental data indicate that the modification process was successful in coating the membrane surface with PEI, which provided amine groups to make the membrane surface positively charged. Although the goal was to increase rejection of bromide ions on the positively charged membrane, the bromide ions concentration increased in the permeate from the PEI-modified membranes. 82% of the bromide ions in the feed were rejected by the PEI modified membrane compared to 94.5% rejection by the unmodified membrane. This behavior can be explained by a combination of two factors, concentration polarization and strength of the electrostatic interactions. The positive charge on the modified membrane surface increased the concentration polarization of bromide ions compared to the case of unmodified membrane. However, the electrostatic attraction forces between bromide ions and PEI may have not been strong enough to retain the bromide ions from diffusion and transport across the membrane. Therefore, modification with PEI was an effective method for enhancing removal of bromide ions by RO membranes. However, the observed increase in water flux for the PEI modified membrane compared to the unmodified one is promising for other applications that are less concerned with disinfection byproducts and their precursors. Furthermore, the results of this study highlights the importance of conducting a more comprehensive membrane performance evaluation for any research on RO membrane modifications because unintended consequences can occur. For example, in the current study, when the membrane was modified, enhanced water flux was observed but the rejection level for some dissolved species was compromised. Therefore, membrane modification research should investigate the membrane performance beyond the goals of the studies to be able to identify

possible side effects that can results from the modifications. As for bromide removal, future research may focus on “embedded” rather than “surface” membrane modifications that can limit diffusion of bromide ions in the membrane or test operational changes that can lead to lower concentration polarization on the membrane surface.

Chapter 2. Systematic Evaluation of Reverse Osmosis Membrane Modifications for Fouling Reduction and Removal of Disinfection Byproducts

2.1 Introduction

Reverse osmosis (RO) membranes are widely used in desalination and advanced wastewater treatment facilities for producing high quality effluents that meet strict regulations [25]. Before distributing treated water to consumers or the environment, harmful pollutants and pathogens have to be removed to an extent that does not pose risks. RO is favored by potable water reuse for facilities for its wide range of targeted pollutants and high rejection [26, 27].

However, RO membranes have drawbacks. During operation, the membranes accumulate soluble inorganic matter, organic matter, particulates, and microbes that form films on the membrane surface, also known as membrane fouling [28]. When soluble inorganic matter adsorbs to the membrane surface, the compounds form scales. The membrane may also be damaged by salt crystal's sharp edges [29]. Fine particulates can form cakes and block the membrane [30]. Molecules with smaller compounds may also block the membrane pores [31]. Thus, fouling has adverse impacts on the energy demand, permeate quality, water flux, and membrane integrity [32, 33]. Not only does fouling increase operation cost, but membranes also have to be replaced when the effluent quality decreases. RO treatment systems may suffer up to 76% decline in flux from fouling [34]. Although water and wastewater pretreatment methods are used to reduce fouling, microbial cells reproduce and need control that is more effective. To reduce fouling, chlorine washing and membrane backwashing are performed. However, biocides like chlorine

can degrade the active polyamide layer of the RO membranes as well as induce further microbial growth by oxidizing large organic molecules to become a food source for microorganisms [35].

Although RO membranes achieve high removal rates for most pollutants, disinfection by-products (DBPs) (e.g., N-nitrosamines) and DBPs precursors (e.g., bromide) have considerably lower removal rates [40, 41]. N-Nitrosodimethylamine (NDMA) are carcinogenic disinfection byproducts found in chlorinated water and wastewater. NDMA was reported to have a rejection of 10% - 40% by RO treatment [57, 58]. This is the lowest compared to RO rejection for other DBPs. For example, RO membrane rejection levels are > 50 % for haloacetonitriles (HANs), > 90 % for haloacetic acids (HAA), and > 60 % for trihalomethanes (THMs) [59-64]. Bromide is a precursor for the formation of bromate (BrO_3^-), which is a carcinogen [42]. In the presence of bromide ions, bromate forms when ozone is used as a water disinfectant or as an oxidizing agent in advanced oxidation processes [42]. Multiple stage RO systems are used to increase DBPs precursor rejection rate. Facilities also rely on UV radiation and dosed hydrogen peroxide to remove disinfection by-products after the precursors react [43]. However, these methods increase energy consumption and may not achieve complete treatment [44].

Virus removal efficiency is a major concern in potable reuse applications [35]. Because the size of viruses is smaller than bacteria, their removal by RO membranes are generally lower than bacteria. Nonetheless, RO membranes still achieve better viruses' removal compared to other removal alternatives [36, 37, 38]. Higher log removal for viruses by RO membranes is desired with the increasing interest in treating water for direct potable reuse. Virus removal can be enhanced by ozone treatment after RO systems [39].

Research on RO membrane modifications has been growing rapidly to improve membrane performance and overcome some of the aforementioned issues (e.g., fouling and removal of DBPs and their precursors). Membrane surface properties such as surface roughness and hydrophilicity have been modified, for example, to reduce adsorption of foulant molecules [65]. By reducing the chances of fouling, membrane maintenance can be minimized. Furthermore, membrane modifications can act as surface barriers to reduce transmission of solutes across the membrane, thus enhancing rejection of unwanted pollutant such as DBPs precursors [67].

Extensive research has been conducted on modifying RO membranes to reduce fouling and increase rejection of water pollutants. No systematic evaluation has been conducted to date to help inform decisions on the most promising modifications. Therefore, the objective of this study was to conduct a multi-criteria decision analysis to systematically evaluate and rank the performance of surface modification alternatives for RO membranes for fouling reduction and all types of improvements for NDMA removal. To achieve this objective, research studies on RO membrane modifications were collected and analyzed. Membrane performance data and testing conditions were extracted from the articles and evaluation criteria for the modification alternatives were identified. Then, a multi-criteria decision analysis was performed by calculating weights for the performance criteria using the analytical hierarchy process (AHP) and using those weights to rank the alternatives using the Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS). After finalizing the ranks, a sensitivity analysis was conducted to evaluate the impact of assumptions made throughout the multi-criteria decision analysis process on the ranks obtained.

The AHP breaks down criteria sets for evaluation and weighs them based on the order of importance [69]. Criteria sets guide the user to select alternative choices that is closest to ideal solution. The AHP has been used to aid decision making for various applications, for example, in treatment design planning and seawater reverse osmosis plant optimization [51, 52]. The main advantages of the AHP include wide applicability, easiness to use, and group decision making. Using the hierarchy system of the AHP helps in complex decision-making processes [53]. The TOPSIS is a popular technique for outranking methods for selection of the best ones. This technique identifies the alternatives that are closest to the positive ideal solution and farthest from the negative ideal solution. The capability of TOPSIS to compare alternatives has been utilized in many applications such as technology planning [54].

2.2 Methodology

2.2.1 Literature Review and Article Selection Process

An online search was conducted to gather research articles on RO membrane modifications for fouling reduction, removal of disinfection by-products and DPBs precursors, and virus removal. The databases used for the search include ScienceDirect, American Chemical Society, and Engineering Village. The articles on membrane fouling reduction were searched using the keywords “fouling” and “reverse osmosis membranes.” One hundred and twenty-two relevant articles were collected. Studies on membrane modifications for the removal of DBPs and precursors were searched and thirty-one relevant articles were found: 26 on NDMA, 2 on bromide, and 3 on other DBPs and their precursors. Because the majority of research on RO

modifications of precursors and DBPs removal focused on NDMA removal, the scope of this review was limited to NDMA.

Research on membrane modifications for virus removal was searched using the keywords "virus" and "reverse osmosis membranes." Only two relevant articles were found and therefore, RO modifications for virus removal was excluded from the multi-criteria decision analysis. Figure 2.1 shows that the majority of the RO membrane modifications research focuses on fouling reduction. This is expected because fouling has been one of the major concerns with the operation of reverse osmosis membrane processes.

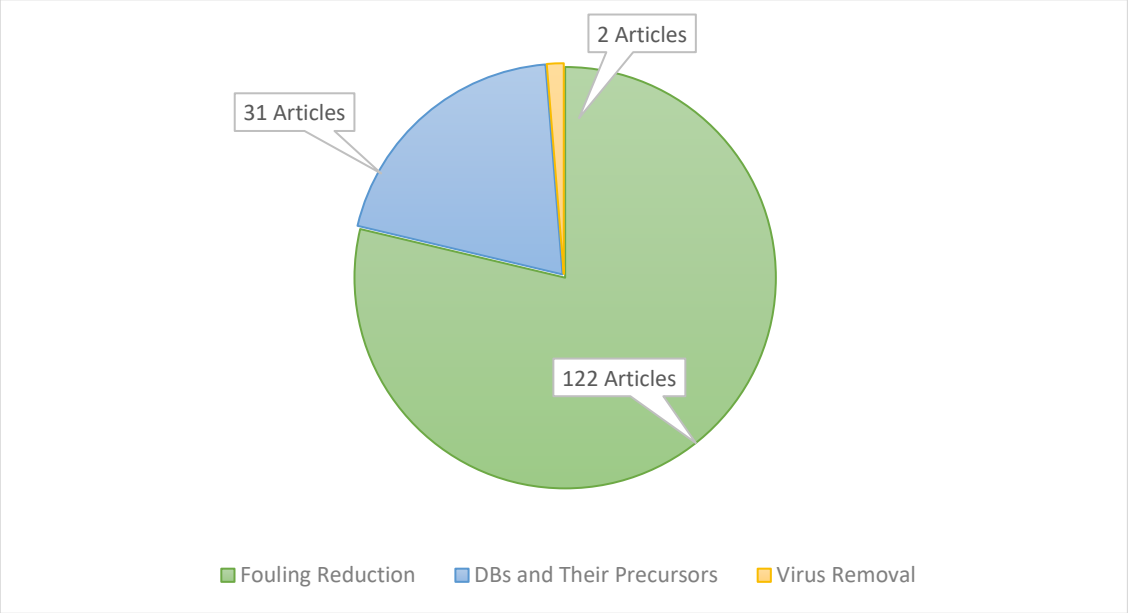


Figure 2. 1 Distribution of articles on reverse osmosis membrane modifications.

The articles collected were classified based on the modification methods and modification objectives (Figure 2.2). The membrane modifications were either surface modifications or embedded modifications (inclusion of modification agents within the membrane structure). The

current study focused only on the modifications made to the RO membrane surface (i.e., embedded modifications were outside the scope of this review and analysis).

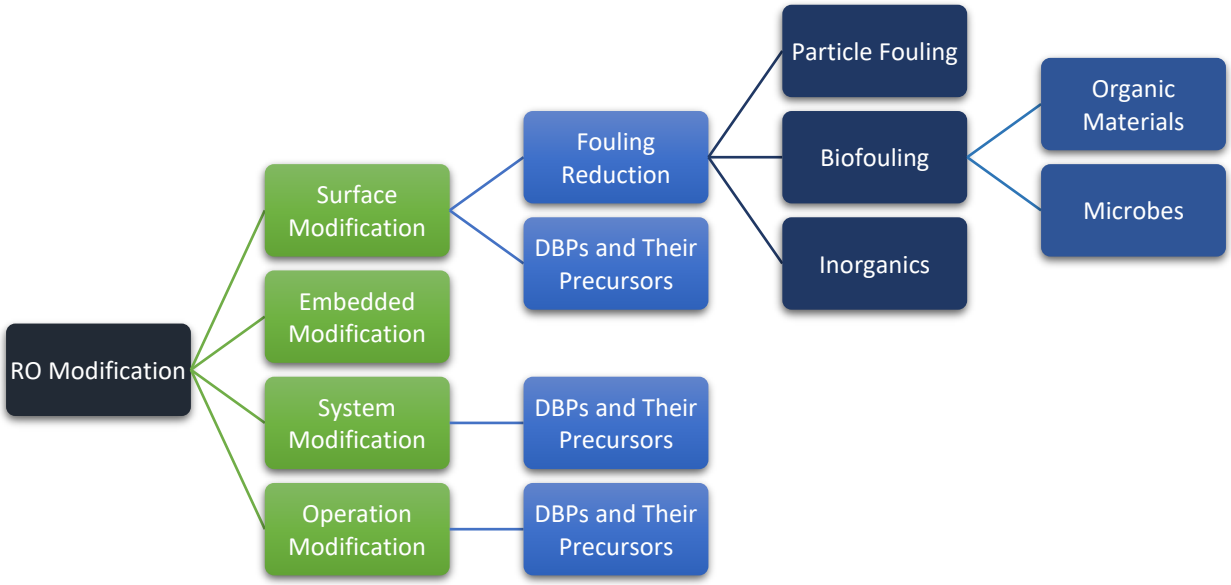


Figure 2. 2 Classification of the studies on RO membrane modifications.

Of the 122 articles on RO surface modifications for fouling reduction, 72 were considered herein for the multi-decision criteria analysis. The decision process for selection of the 72 articles on fouling reduction is presented in Figure 2.3. For NDMA removal, 13 out of the 26 collected articles were excluded from this analysis because they were not experimental (i.e., theoretical analysis for prediction of RO membrane performance). The 12 remaining articles on NDMA removal were not only related to RO membrane surface modifications, 3 articles modified the RO system setup and 3 articles made operational modifications for enhancing the removal of NDMA. Due to the limited number of studies on NDMA removal enhancement using surface modifications, all of the 12 articles on NDMA removal were considered in the multi-criteria decision analysis. Thus, the scope of the analysis was not only limited to surface modifications to

the RO membranes, rather it was expanded to encompass any type of modification that enhances NDMA removal.

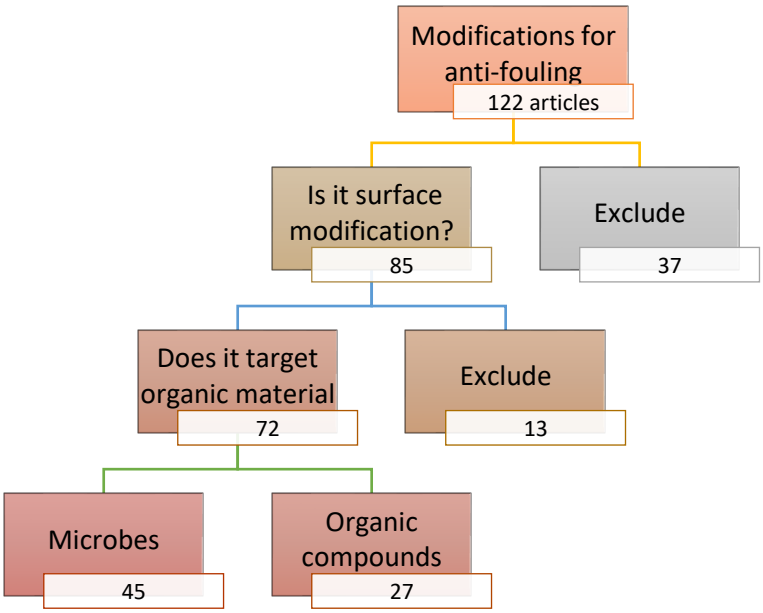


Figure 2. 3 Decision process for selection of the studies on fouling reduction.

Performance data on fouling reduction and NDMA removal as well as information on operational conditions and the modification methods were gathered from the selected articles (72 on fouling and 12 on removal of NDMA). This information was tabulated and statistics were extracted on the types of modifiers used, modification approaches, foulants targeted, scale of the studies, performance metrics, and more.

2.2.2 Multi-Criteria Decision Analysis (MCDA)

Multi-criteria decision analysis (MCDA) was conducted to prioritize research on modifications for fouling reduction and enhancement of NDMA removal. For fouling reduction, only RO membrane surface modifications studies were considered for the MCDA. One the other hand, the

MCDA for enhancing NDMA removal included studies using all types of modifications. The AHP and TOPSIS processes were collectively used for ranking the modification alternatives based on the evaluation criteria presented in Table 2.1.

Table 2. 1 The evaluation criteria for ranking alternative modifications

Modification Goal	**Evaluation Criteria**
Fouling Reduction	Compared to unmodified membranes: • Flux difference in the absence of foulant* • Flux difference after fouling occurred* • Salt removal difference in the absence of foulant* • Feed salt concentration
NDMA Removal Enhancement	• Rejection of NDMA • Pressure demand • Improvement approach - Operation modification - Membrane modification - System modification

* The "difference" was calculated by subtracting the values (e.g. flux or salt removal) of the unmodified membranes from those of the modified membranes.

Using the AHP, a pairwise comparison was conducted to weigh the evaluation criteria based on their relative importance. A value between 1 and 4 was assigned for each criterion, 1 being equally important and 4 being strongly more important. The criteria were arranged in a matrix and the values in the rows were divided by those in the columns to calculate indicators of relative importance for the evaluation criteria for fouling reduction (Table 2.2) and NDMA removal (Table 2.3). For example, the flux difference after fouling occurred was considered 4 times more important than the feed salt concentration (Table 2.2).

Table 2. 2 Pairwise comparison matrix for fouling reduction

Modification Type	**Flux difference in the absence of foulant**	**Flux difference after fouling occurred**	**Salt removal difference in the absence of foulant**	**Feed salt concentration**
Flux difference in the absence of foulant	1.00	0.50	1.00	2.00
Flux difference after fouling occurred	2.00	1.00	2.00	4.00
Salt removal difference in the absence of foulant	1.00	0.50	1.00	2.00
Feed salt concentration	0.50	0.25	0.50	1.00

Table 2. 3 Pairwise comparison matrix for NDMA removal

	Rejection of NDMA	**Pressure**	**Improvement**
Rejection of NDMA	1.00	1.50	3.00
Pressure	0.67	1.00	2.00
Modification Type	0.33	0.50	1.00

The sum of each column in Table 2.2 and Table 2.3 was determined and the value in each cell was divided by the sum of the respective column. The weight of each evaluation criterion was then calculated by averaging the values of each row. To limit bias in the weighting process, a consistency ratio < 0.1 needs to be achieved. The consistency ratio was calculated by dividing the consistency index by the random index (Equation 1).

$$Consistency\ Ratio = \frac{Consistency\ Index\ (C.I.)}{Random\ Index\ (R.I.)} \quad (1)$$

The consistency index (C.I) was calculated using Equation 2.

$$Consistency\ Index\ (C.I.) = \frac{\lambda_{max} - n}{n-1} \qquad (2)$$

Where, λ_{max} is the weighted sum value of criteria weights normalized by the sum of indicators of relative importance of each criterion and n is the number of criteria. The C.I. was then divided by a random index, which was determined by the number of criteria used in AHP, to calculate the consistency ratio.

The TOPSIS analysis was performed using the criteria weights obtained from the AHP. In the TOPSIS matrix, the value for each evaluation criterion was the actual data reported in the articles for that criterion. For non-numeric criteria, the linguistic values were converted to numbers based on a scale of 1 to 3. The data in the TOPSIS matrix were normalized using Equation 3.

$$\bar{X}_{ij} = \frac{X_{ij}}{\sqrt{\sum_{j=1}^{n} X_{ij}^2}} \qquad (3)$$

Where, X_{ij} is the row element and n is the number of elements in the row.

The normalized data determined using Equation 3 were multiplied by their respective criteria weights to calculate V_{ij}, which is the normalized value of the element. Then, the highest

normalized value (V_j^+) and the lowest normalized value (V_j^-) were identified and used for calculating the Euclidean distance from the ideal solutions (S_i^+ and S_i^-) using Equations 4 and 5.

$$S_i^+ = [\sum_{j=1}^{m}(V_{ij} - V_j^+)^2]^{0.5} \quad (4)$$

$$S_i^- = [\sum_{j=1}^{m}(V_{ij} - V_j^-)^2]^{0.5} \quad (5)$$

Where, (V_j^+) is the positive ideal solution, (V_j^-) is the negative ideal solution, S_i^+ is the Euclidean distance from the ideal best solution, and S_i^- is the Euclidean distance from the ideal worst solution. It is noted that the ideal positive solution is the alternative with the best possible outcome based on the evaluation criteria.

Using the Euclidean distance values, a performance index (P_i) was calculated using Equation 6. The performance index (P_i) values were used for ranking the modification alternatives (higher P_i indicates higher rank).

$$P_i = \frac{S_i^-}{S_i^+ + S_i^-} \quad (6)$$

2.2.3 Sensitivity Analysis

A sensitivity analysis was performed to evaluate the impact of changing the weights of the evaluation criteria and the values assigned for unreported data on the MCDA ranking results. The weight obtained using the AHP for each evaluation criterion was increased by 20% in 5%

increments while the weights of the remaining criteria were decreased by the same percent to maintain a total weight of 100%. The Pi values were re-calculated each time the weight of a criterion changed. The new ranking for the alternatives was determined and compared to the original ranking obtained using the AHP weights to assess the sensitivity of the ranking results to any judgment bias or assumptions made to perform the MCDA.

2.3 Results and Discussion

2.3.1 Analysis of the Literature on RO Modifications

Eighty-four articles on RO modifications were analyzed, 86% on fouling reduction and 14% on NDMA removal (Figure 2.4). To analyze the efficacy of RO modification approaches, the articles were searched for the following information: water flux, salt rejection, fouling reduction, NDMA removal, feed salt concentration, potential toxicity of the materials used for modification, amounts of materials used for modification, cost and scalability of the modification method, longevity of the modification, membrane type, and life cycle of the membranes. The information found on these performance criteria and testing conditions were tabulated and further analyzed as presented in Sections 3.1.1 and 3.1.2. It is noted that less than 40% of the aforementioned criteria were reported in any of the articles reviewed. This highlights future research needs to fill gaps in the literature to better inform decisions on the efficacy and practicality of RO membrane modifications approaches.

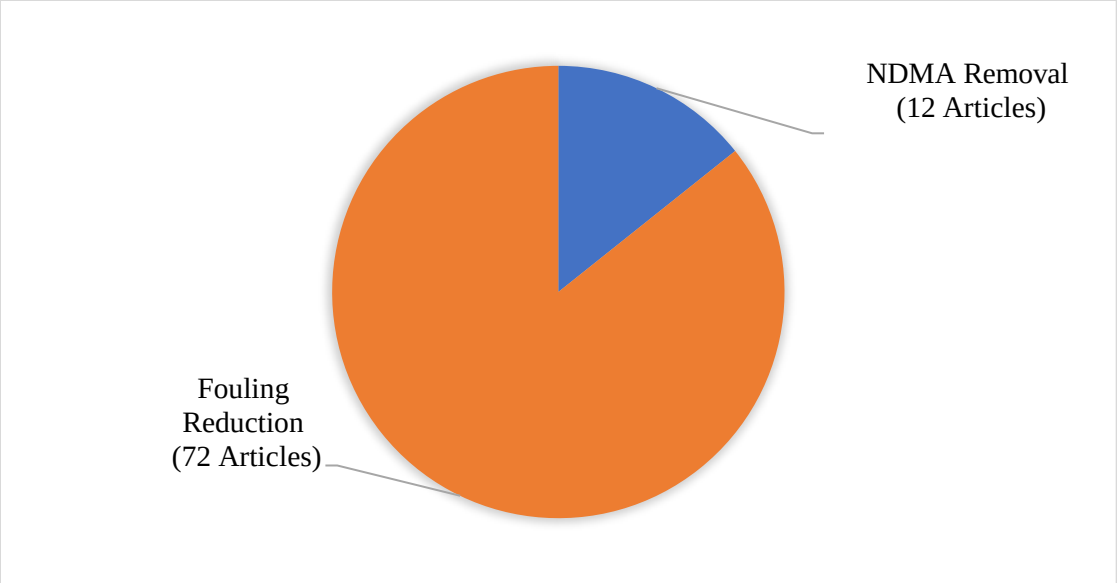


Figure 2. 4 Distribution of the articles selected for analysis.

2.3.1.1 Fouling Reduction

The articles on membrane fouling were classified based on the method used for surface modification to 14 categories (Figure 2.5). Grafting and coating were the dominant RO membrane surface modification methods. In general, these two methods require less equipment and are less complicated to perform compared to the other ones listed in Figure 2.5 [149]. Grafting modifies RO membranes by imparting chemical functional groups on their surfaces [150]. The grafting can be categorized into "grafting from" and "grafting to" methods [150]. During the "grafting from" methods, functional groups become polymerized onto the surface through a chain propagation reaction [150]. During the "grafting to" methods, target compounds are prefabricated before attaching to membrane surface [150]. Coating on the other hand is performed by dispersing an aqueous polymer solution on the membrane surface and a film is created on the surface once the solvent evaporates [149]. Grafting and coating are typically used

for increasing membrane hydrophilicity; however, they also reduce permeate flux to varying degrees [150].

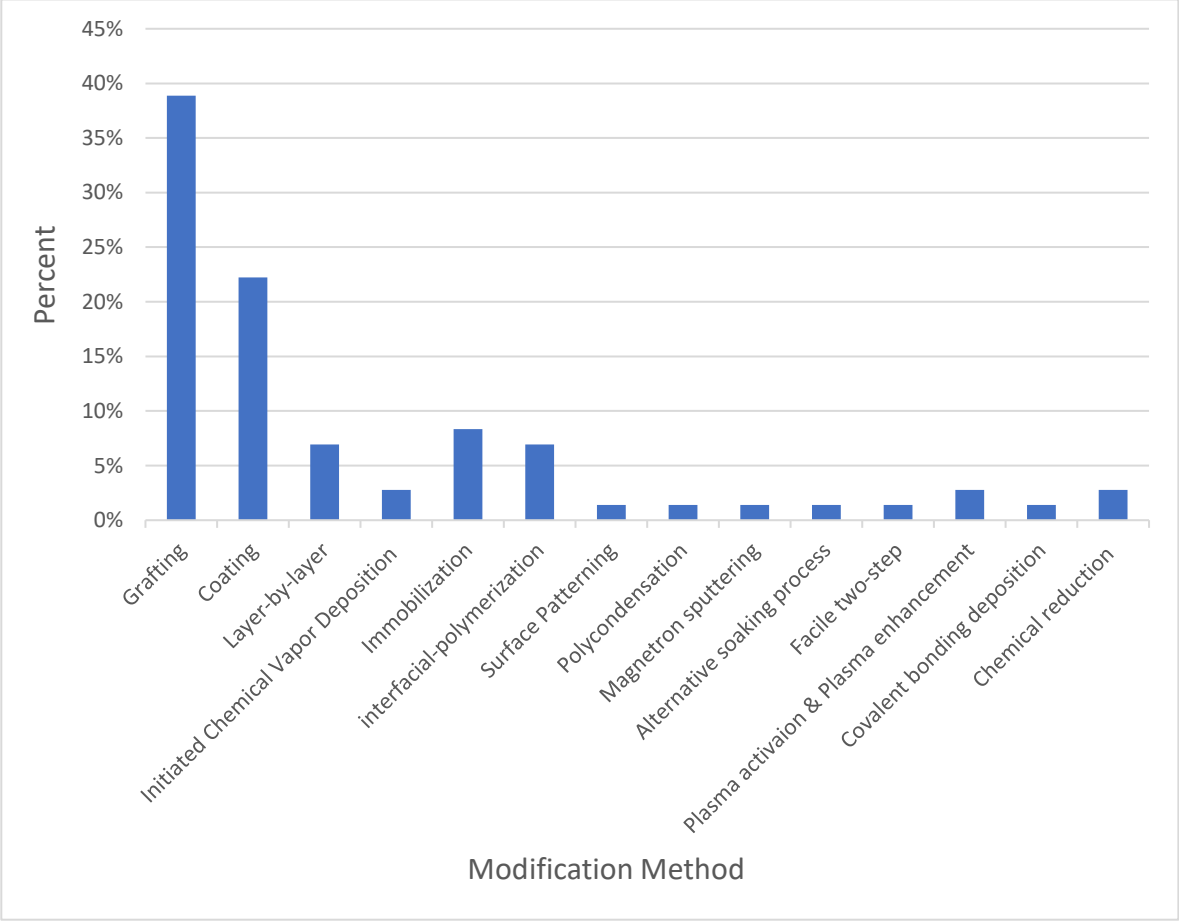


Figure 2. 5 Distribution of articles based on membrane modification methods.

Each of the modification methods presented in Figure 2.5 involves the use of different types of chemical modifiers. The dominant ones used for reducing fouling are polymers (Figure 2.6). This is likely because most RO membranes are synthesized from polymeric materials and thus, polymers are compatible modifiers. Examples of the polymers used include hydrogen bond-donating groups such as polyethylene glycol (PEG), biocidal polymers such as biomimetic polydopamine (PDA), and antimicrobial polymers such as guanidine-based polymer [150, 151].

Nanoparticles were the second mostly utilized materials for RO membrane surface modifications. Commonly used nanoparticles include titanium dioxide (TiO_2), zinc oxide (ZnO), magnesium oxide (MgO), and silver nanoparticles (AgNPs) [151]. These nanoparticles were mainly used to reduce biofouling because of their antimicrobial properties [150]. Zwitterionic materials have a net charge of zero and include negatively and positively charged atoms joined by covalent bonds [150]. Examples of these materials include polysulfobetaine, carboxybetains, and 1-cysteine. Zwitterionic materials are favorable for marine anti-biofouling applications because of their high resistivity to proteins and bacterial films [150]. Graphene oxide is obtained by reacting graphite with oxidizers. Graphene has been studied for desalination technologies for its tensile strength, permeability resistant of small molecules, and negligible thickness [153]. The graphene oxide sheets can also have nanoscale pores embedded to improve surface hydrophilicity, which enhances antifouling properties [153]. The "Other" category in Figure 2.6 includes metal carbides ($Ti_3C_2T_x$) and polyamides (2-aminoimidazole).

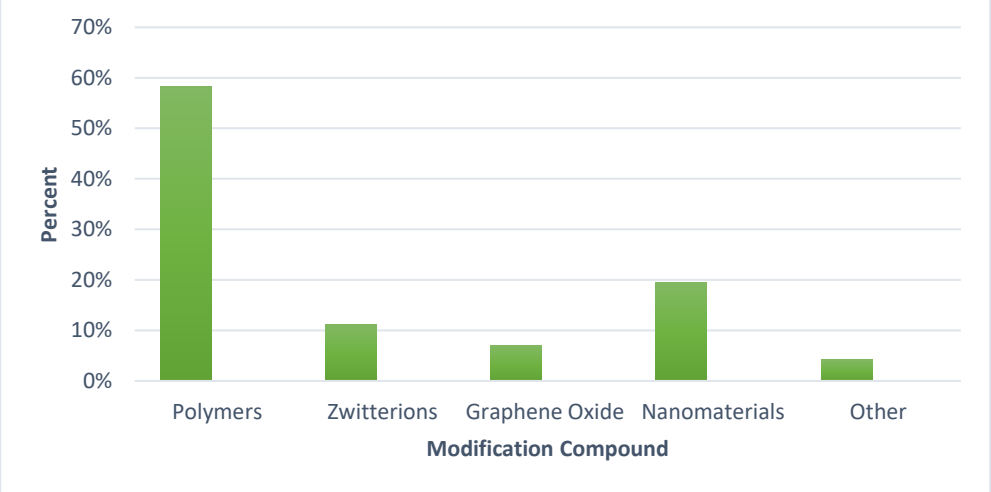


Figure 2. 6 Distribution of articles based on the modification compound used.

Organic compounds and microbes are a major cause of RO membrane fouling [154]. Around 60% of the membrane surface modification studies focused on reducing organic fouling and the remaining 40% focused on reducing microbial fouling. Bovine serum albumin (BSA), a protein derived from cows, was the organic fouling agent commonly used for studying the effectiveness of surface modified RO membranes against fouling. For reduction in microbial fouling, research investigations tested the growth inhibition of bacteria that are typically present in wastewater and freshwater streams with 56% of the studies using E. coli as a model microbial foulant (Figure 2.7).

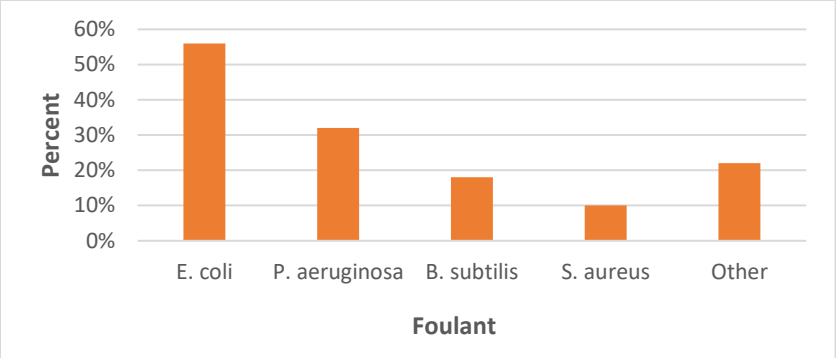


Figure 2. 7 Distribution of articles based on microbial foulants tested.

Four evaluation criteria were used to assess the effectiveness of the RO membrane surface modification alternatives for reducing fouling. These criteria were the water flux difference between the modified and unmodified membranes in the absence of foulants, water flux difference in the presence of foulants, salt removal difference in the absence of foulants, and feed salt concentration. The first three criteria were calculated based on information gathered from the articles reviewed. Table 2.4 presents the data for the four evaluation criteria used to assess fouling reductions because of RO membrane surface modifications.

Table 2. 4 Performance data for RO membrane modifications to reduce fouling

Modification Type	Description	Foulant	Goal	Feed salt concentration	Flux difference in the absence of foulants (LMH)	Flux difference after fouling (%)*	Salt removal difference (%)
iCVD	zwitterionic copolymer was synthesized	BSA	organic	2000 ppm NaCl	-42	122	2
LbL	FeCL3 at different phytic acid assemply	BSA	organic	2000 ppm NaCl	-10	162	1
interfacial polymerization	Ti3C2Tx was used to modify the membranes	BSA	organic	2000 ppm NaCl	0	136	-1
coating	zwitterionic sulfobetaine silane compounds EPBS and MPPS	Shewanella, Vibrio, Oceani monas, Pseudoalteromonas.	microbial	37400 ppm NaCl	NR	NR	-1
coating	polydopaminewith Trizma hydrochloride was used to modify	BSA	organic	2000 ppm NaCl	7	116	0
polycondensation	poly(guanidine-hexamethylenediamine-PEI) (poly(GHPEI)) was used	E.coli and Bacillus subtilis	microbial	2000 ppm NaCl	0	236	2
coating	sericin-saturated membrane	BSA	organic	500 ppm NaCl	-2	143	1
interfacial polymerization	Glutaradehyde (GA, 25 % aqueous solution) and 3-amino-1-propanol (AP) and ethylenediamine (EDA)	Escherichia coli	microbial	2000 ppm NaCl	19	123	-1
interfacial polymerization	pristine multi-walled carbon nanotubes w/ ammonium persulfate and p-toluenesulfonic acid (MWCNTs-PPY)	BSA	organic	2000 ppm NaCl	12	125	1
magnetron sputtering	plasma activated and deposited silver nano particles	pseudomonas	microbial	2000 ppm NaCl	17	NR	0

Modification Type	Description	Foulant	Goal	Feed salt concentration	Flux difference in the absence of foulants (LMH)	Flux difference after fouling (%)*	Salt removal difference (%)
interfacial polymerization	Ce (IV)-induced graft polymerization of MSA from GO-NH2 (GO-g-PMSA)	BSA	organic	1000 ppm NaCl	8	133	4
grafting	PEI	lysozyme	organic	1500 ppm NaCl	-25	175	1
chemical grafting	ε-Poly-l-lysine (PL) polypeptide	BSA	organic	2000 ppm NaCl	12	116	-1
grafting	zwitterionic polymer carboxybetaine methacrylate (CBMA)	B. subtilis E. coli	microbial	2000 ppm NaCl	15	165	0
grafting	[3-(Methacryloylamino)propyl]dimethyl (3-sulfopropyl)ammonium hydroxide inner salt (3-SBMA)	EPS	organic	36,000 ppm NaCl	12	112	-1
grafting	zwitterionic surface from l-cysteine, amino and carboxylate groups of l-cysteine	BSA	organic	2500 ppm NaCl	0	127	3
LbL	Tobramycin (TOB) bilayer with poly acrylic acid (PAA)	B. subtilis E. coli	microbial	2000 ppm NaCl	10	228	1
interfacial polymerization	p-aminophenol modified GO on surface	E. coli and S. aureus	microbial	2000 ppm NaCl	3	NR	0
grafting	N-isopropylacrylamide followed by acrylic acid	BSA	organic	500 ppm NaCl	-8	151	2
coating	polyvinyl alcohol (PVA) and MPTES	BSA	organic	2000 ppm NaCl	-2	149	1
coating	3-mercaptopropyltrimethoxysilane (MPTES) and PVA	BSA	organic	32,800 ppm	30	NR	29

Modification Type	Description	Foulant	Goal	Feed salt concentration	Flux difference in the absence of foulants (LMH)	Flux difference after fouling (%)*	Salt removal difference (%)
grafting	anionic 2-carboxyethyl acrylate (CAA) and cationic [2-(acryloyloxy)ethyl] trimethyl ammonium chloride (TMA)	BSA	organic	7650 ppm NaCl	-1	263	-3
alternative soaking process	BaSO4 based coating	BSA	organic	500 ppm NaCl	6	105	2
facile two-step	poly(N-vinylpyrrolidone) (PVP)	BSA	organic	2000 ppm NaCl	-5	134	0
plasma activation	zwitterionic SBMA	Pseudomonas	microbial	2000 ppm NaCl	45	NR	0
iCVD	zwitterionic ammonium hydroxide (SPE)	Pseudomonas putida	microbial	10 ppm NaCl	0	140	NR
coating	Sulfonated polyvinyl alcohol (SPVA) and compared to polyvinyl alcohol (PVA)	BSA	organic	2000 ppm NaCl	-10	148	1
SC	P(NIPAM-co-Am)	BSA	organic	2000 ppm NaCl	5	110	0
SG	l-cysteine	BSA	organic	2500 ppm NaCl	-2	128	3
SG	NIPAm, AA	BSA	organic	500 ppm NaCl	0	156	-1
SC	PVA, PHMG	P. aeruginosa, E. Coli, and B. subtilis	microbial	10 ppm NaCl	72	NR	1
SC	HEMA, PFA	E. Coli	microbial	200 ppm NaCl	-17	153	4
SG	pSBMA	BSA	organic	2000 ppm NaCl	0	116	0
SG	ADMH, MBA	E. Coli	microbial	2000 ppm NaCl	-16	NR	0

Modification Type	Description	Foulant	Goal	Feed salt concentration	Flux difference in the absence of foulants (LMH)	Flux difference after fouling (%)*	Salt removal difference (%)
SC, SG	PDA, BiBBr, MTAC	Alphaproteobacteria and Gammaproteobacteria	microbial	2000 ppm NaCl	8	116	9
SC	p(MDBAC-r-Am-r-HEMA)	BSA	organic	2000 ppm NaCl	-59	NR	1
SC	p(MPC-co-AEMA)	Sphingomonas paucimobilis	microbial	5000 ppm NaCl	0	NR	2
SC	HPOEM, PEI	BSA	organic	32,000 ppm NaCl	-1	95	0
SC	AUTEAB	E. Coli	microbial	100 ppm NaCl	15	113	NR
SG	PVA	BSA	organic	500 ppm NaCl	0	140	1
SC	p(4-VP-co-EGDA), pCBAA	E. Coli and B. subtilis	microbial	2000 ppm NaCl	-7	92	2
SG	DMAEMA, CBMA	E. Coli and Bacillus subtilis	microbial	2000 ppm NaCl	15	124	0
SG	PEI	lysozyme	organic	2000 ppm NaCl	-15	180	1
SG	GPPTMS	caesin	organic	2000 ppm NaCl	NR	NR	1
Chemical reduction	AgNP	E. coli	microbial	50 ppm NaCl	-11	NR	0
		P. aeruginosa	microbial				
		S. aureus	microbial				

Modification Type	Description	Foulant	Goal	Feed salt concentration	Flux difference in the absence of foulants (LMH)	Flux difference after fouling (%)*	Salt removal difference (%)
Chemical reduction	CuNP	E. coli	microbial	50 ppm NaCl	13	NR	-1
Dip coating	Iron NPs on graphene oxide	B. halotolerans	microbial	2000 ppm NaCl	-4	160	NR
Plasma-enhanced magnetron sputtering deposition	AgNP	P. fluorescen	microbial	2000 ppm NaCl	2	NR	0
LbL	PAA/TOB	E. coli, B. subtilis	microbial	2000 ppm NaCl	8	198	0
LBL	Chlorhexidine/glutaladehyde	P. aeruginosa	microbial	100 ppm NaCl	-2	196	0
LbL	TiO2/GO	E. coli	microbial	1000 ppm NaCl	4	198	0
Grafting	pMEDSAH	S. paucimobilis	microbial	850 ppm NaCl	-3	353	-1
Grafting	MPC	P. fluorescen	microbial	2000 ppm NaCl	1	NR	0
Grafting	Poly(sulfobetaine)	Sodium alginate	microbial	2000 ppm NaCl	10	NR	0
Grafting	NIPAM and ZnO nanoparticles	E. coli	microbial	2000 ppm NaCl	3	264	1
Grafting	PANI/CuNP	E. coli	microbial	2000 ppm NaCl	4	49	1
Covalent bonding and deposition	Cysteamine/CuNP	E. coli	microbial	50 ppm NaCl	NR	NR	NR

Modification Type	Description	Foulant	Goal	Feed salt concentration	Flux difference in the absence of foulants (LMH)	Flux difference after fouling (%)*	Salt removal difference (%)
Grafting	Chitosan/CuNP	E. coli	microbial	2000 ppm NaCl	-13	133	0
Grafting	2-aminoimidazoles	P. aeruginosa	microbial	500 ppm NaCl	NR	NR	1
Grafting	TC	E. coli	microbial	2000 ppm NaCl	0	122	1
		P. aeruginosa					
		S. aureus					
Grafting	poly(3-allyl-5,5-dimethylhydantoin-*co*-vinylamine) (P (ADMH-co-Vam))	E. coli	microbial	2000 ppm NaCl	5	212	1
Grafting: Michael-type addition and Schiff base reaction	PDA-modified membrane was further covered by poly(GHPEI)	E. coli	microbial	2000 ppm NaCl	-7	353	1
		B. subtilis	microbial				
Grafting: Ag–S	Cysteamine modified TFC-AgNP/SiO2	E. coli	microbial	2000 ppm NaCl	0	NR	0
		P. aeruginosa					
		S. aureus					
Grafting: UV-assisted	RWRWRWA-(Bpa) peptide	P. aeruginosa	microbial	100 ppm NaCl	-13	NR	6
Immobilization	PPy	E. coli	microbial	2000 ppm NaCl	12	13	0
Immobilization	Melamine and PEG	E. coli ad B. subtilis	microbial	2000 ppm NaCl	24	160	1
Immobilization	AgNP	E. coli	microbial	2000 ppm NaCl	20	NR	-1
		P. aeruginosa					
		S. aureus,					

Modification Type	Description	Foulant	Goal	Feed salt concentration	Flux difference in the absence of foulants (LMH)	Flux difference after fouling (%)*	Salt removal difference (%)
immobilization	GO nanosheet	P. aeruginosa	microbial	2000 ppm NaCl	-25	175	5
Immobilization substrate and PA layer	GO nanosheet	E. coli cells	microbial	50 ppm NaCl	8	NR	16
Immobilization :PA layer	Pal/TiO2	E. coli	microbial	2000 ppm NaCl	20	20	0
Surface patterning: sol−gel based nanoimprinting	TiO2 pillar	P. aeruginosa	microbial	2000 ppm NaCl	-19	130	0

*Flux difference after fouling was calculated by subtracting normalized percent of modified membrane flux by normalized percent of unmodified membrane flux. The normalized percent was calculated by dividing flux after fouling by flux before fouling. A percentage below 100% indicates reduction in flux difference after fouling.

The flux difference in the absence of fouling was used as an indicator for whether or not the surface modifications changed the permeability of the membranes. After fouling occurs, the flux difference between the modified and the unmodified membranes was used as an indicator for the efficiency of modifications to reduce fouling and maintain water flux. Greater flux difference suggests less fouling. Salt removal difference between the modified and unmodified membranes was also considered to assess whether the membrane modifications resulted in compromising the membranes' efficiency to reject salts.

The membrane modification studies on fouling reported a wide range of water flux which could be explained by the variability in testing conditions. For example, the operating pressures used in the studies varied between 1 to 27.6 bar and in general, increasing pressure results in an increase in water flux. The water flux ranged from 50 to 100 $L/m^2 h$ in the absence of fouling for modified and unmodified membranes. After fouling occurred, the water flux ranged mostly from 15 to 70 $L/m^2 h$.

When surface-modified membranes were used, approximately 50% of the studies reported a reduction in water flux compared to unmodified membranes in the absence of fouling. The flux reduction was less than 25 L/m^2h; however, drastically larger reductions (e.g., 59 L/m^2h) were also observed (Table 2.4). The flux reduction after modification was assumed to result from the decrease in porosity and alteration of hydrophilicity of the modified membrane [149]. However, after fouling occurred, <10% of the studies reported a reduction in water flux from the modified membranes as compared to the unmodified ones. This indicates that these studies were not successful in reducing fouling.

Fouling reduction was also assessed by dividing the water flux after fouling occurred by the flux before fouling. This ratio was calculated for both the modified and unmodified membranes and was used as an indicator for the changes in membrane permeability due to fouling. These findings suggest that the majority of surface modification methods were successful in improving the fouling resistivity of the RO membranes without compromising the permeate flux. Approximately 90% of the studies maintained salt rejection levels greater than 90% after membrane modifications. Only 10 studies showed reduction in salt rejection after modification.

The majority (64%) of the fouling studies were conducted using feed salt concentration of 2000 mg/L sodium chloride. Approximately 6% of the studies experimented with salt concentrations > 30,000 mg/L, 16% used salt concentration between 2000 - 7650 mg/L, and 14% of the studies used feed salt concentration below 1000 mg/L in 14% of the studies. Salt concentration in the feed water is an indicator for the ability of the membranes to maintain their rejection efficiency in the presence of high salt concentrations. High feed salt concentrations are typically associated with a decrease in salt rejection efficiency of RO membranes [97]. The majority of the fouling studies reported salt rejection above 90% for modified and unmodified membranes in high salt concentrations as well.

2.3.1.2 Removal of NDMA

Improvement in the removal of NDMA was achieved by one of three approaches, surface modifications of the RO membrane, operational modifications, or system modification (~ 25% of the studies) (Figure 2.8). For operational modifications, the removal of NDMA was improved by

changing the pressure, temperature, or pH of the water. System modifications refer to changes in the configuration of the RO system, for example, placing the pressure vessels in series, in parallel, or using a tapered design to enhance removal of NDMA [67]. It is noted that ~ 50% of the studies on the removal of NDMA were pilot and industrial scale, unlike the fouling reduction studies, which were mainly lab-scale.

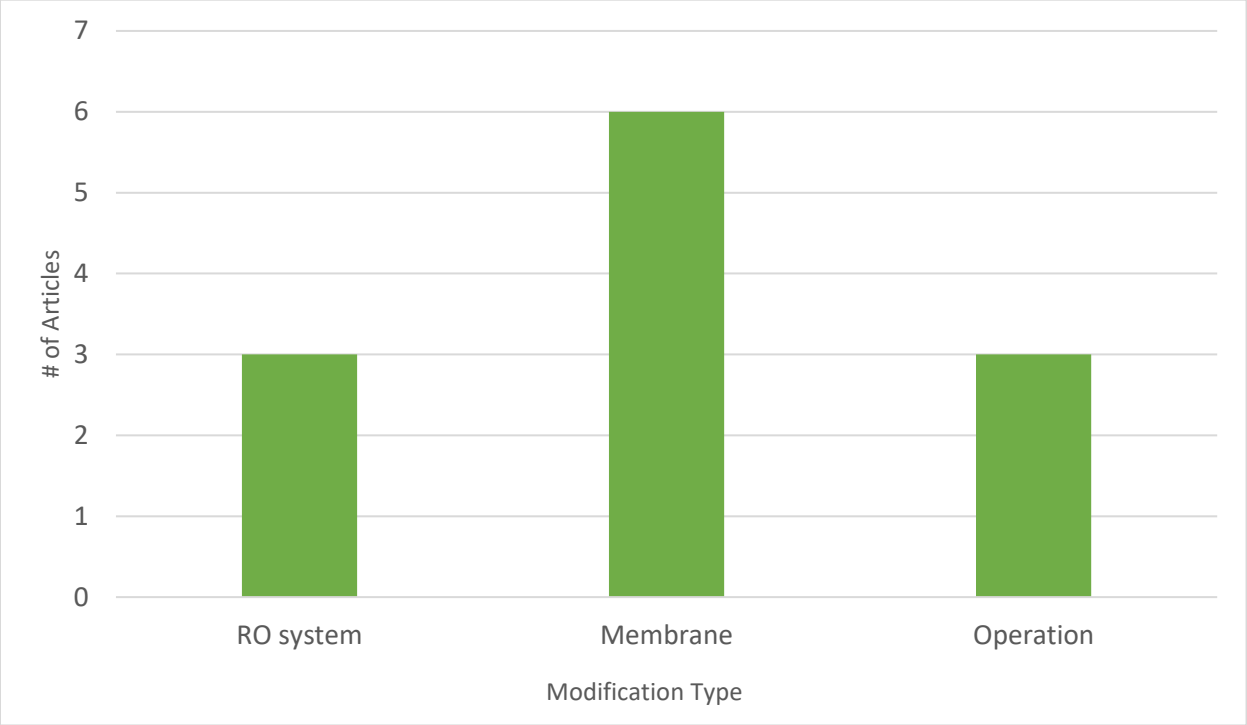


Figure 2. 8 Distribution of articles based on the modification approach for NDMA removal.

The rejection of NDMA ranged between 48 to 98%, and about half of rejection percentages were above 80% (Table 2.5). The pressure used for testing ranged from 145 to 588 psi. About half of the studies used pressures less than 200 psi. In general, the increase in pressure results in increase in rejection of pollutants by RO membranes [63]. Achieving high NDMA rejection while using

lower pressure was used herein as an indicator for the effectiveness of the modification approach. For example, research on heat-treated HYDRApro501 membranes achieved 98% NDMA removal at a relatively low pressure (223 psi) [63]. About 67% of the studies used a coating approach to modify the RO membranes. For example, Croll et al., coated the membranes with graphene oxide to reduce the surface porosity and consequently increase the rejection of NDMA [66]. Besides coating, other membrane modifications approaches for enhancing NDMA removal included heat-treatment and chemical cleaning of the membranes. Heat-treatment was achieved by immersing RO membranes in ultrapure water in high temperature (80 °C) [68]. The treatment tightens the membrane structure, reduces water permeability, and improves rejection of hydrophilic compounds such as NDMA [68]. For chemical cleaning methods, membranes were immersed in a container with a cleaning solution (e.g., NaOH, HCL, or citric acid) [72]. The study was aimed to assess the impact of increased membrane exposure to such chemicals on rejection of NDMA.

Table 2. 5 Industrial and lab scale studies on RO modification for NDMA removal

Description	Improvements	NDMA Removal (%)	Pressure (Psi)	Scale
Performance of multi-staged RO setup	multi-stage process	92	588	Industrial
Multistage RO system study using modeling analysis	spiral round multi-stage	90	565	Industrial
Coating PVA active skin layer on ESPA2 to make LFC3 membrane	coating PVA	40	145	Industrial
Study of performance of multi-staged RO setup in facilities	multi-stage process	58	588	Industrial
TFC-HR and ESPA2 were used to analyze cleaning chemicals' effect on NDMA removal	chemical cleaning	42	145	Industrial
Simulation of full-scale RO system	operation	49	147	Industrial
ESPA3, LFC3, and BW30 dip-coated with polyether-polyamide block copolymer	coating	55	225	Lab
SW30HR with graphene oxide(GO) by tethering to polyamide	coating	82.7	400	Lab
ESPA2, ESPAB, and HYDRApro501 membranes	heat-treat	98	223	Lab
Various RO and UF membranes analyzed for NDMA removal	coating	80	145	Lab
NF90, SWC5, and TFC-HR membranes	operation	80	145	Lab
Low pressure RO membrane system modeling	operation	60	147	Lab

2.3.2 Multi-Criteria Decision Analysis

The AHP and TOPSIS methods were used to rank the modification alternatives to inform decisions on the most promising ones. To achieve that, a number of evaluation criteria were selected, and their order of importance was determined based on the modification goal (e.g., fouling reduction or NDMA removal).

2.3.2.1 Fouling Reduction

Increased resistance to fouling, consistency of permeate flux over the fouling period, and high salt rejection were used herein as indictors for effective membrane modifications to reduce fouling. These indicators were translated to the following performance evaluation criteria: 1) difference in flux between modified and unmodified membranes in the absence of foulant, 2) difference in flux between modified and unmodified membranes after fouling occurred, and 3) salt removal difference between modified and unmodified membranes in the absence of foulant. In addition to these three criteria, salt concentration of the feed water was included as a criterion in the MCDA. Although not being a fouling performance indicator, feed salt can reduce the membrane performance at high concentrations. Thus, it was used as a criterion in the MCDA to compare alternative modifications.

The flux difference after fouling occurred was assigned the highest relative importance in the AHP evaluation. Lower flux difference indicates increased fouling. Then, flux difference and salt removal difference in the absence of foulant were both considered second in the level of

importance, and feed salt concentration was considered the least important among the evaluation criteria. Considering this ranking of importance, the weights for the evaluation criteria calculated using AHP are presented in Table 2.6. The AHP met the limit of consistency (<0.1). Therefore, the AHP weights were used in the subsequent TOPSIS process to calculate performance indexes to rank the modification alternatives.

Table 2. 6 Weights of the evaluation criteria for fouling reduction

Evaluation Criteria	**Weight**
Influent salt concentration	11.1%
Flux difference in the absence of foulant	22.2%
Flux difference after fouling occurred	44.4%
Salt removal difference in the absence of foulant	22.2%

The performances indexes (with top 4 over 0.8 Pi) for the alternative modification methods as using the TOPSIS approach. The top five membrane modifications, based on the values of the Pi, are presented in Table 2.7. Grafting and coating were the methods used for modifying the top 5 alternatives. The top 4 modifications were tested under high feed salt concentration (>30,000 ppm).

Table 2. 7 Top 5 RO membrane modifications for fouling reduction

Rank	RO Modification Method	Pi
1	Grafting with 3-SBMA	0.88
2	Coating with zwitterionic sulfobetaine silane compounds	0.87
3	Coating with MPTES and PVA	0.84
4	Coating with p(MDBAC-r-Am-r-HEMA)	0.82
5	Grafting with anionic 2-carboxylethyl acrylate	0.28

Grafting with [3-(Methacryloylamino)propyl]dimethyl(3-sulfopropyl)ammonium hydroxide inner salt (3-SBMA) was ranked highest with a Pi of 0.88. This modification maintained the level of water flux after fouling occurred compared to an 11% drop in water flux after fouling occurred for the unmodified membrane. Although salt removal difference in the absence of fouling dropped by 1% after modification, the membrane grafted with 3-SBMA maintained relatively high salt rejection (98%). It is noted that the 3-SBMA membrane performance was tested using a high feed salt concentration, which is another indicator for the effectiveness of this modification alternative.

Coating with zwitterionic sulfobetaine silane compounds ranked second with a Pi of 0.87. Although this modification also maintained high salt rejection 99%, flux difference in the absence of foulant was not reported, resulting in a slightly lower Pi than grafting with 3-SBMA. Coating with 3-mercaptopropyltrimethoxysilane (MPTES) and polyvinyl alcohol (PVA) ranked third with a Pi value of 0.84. The salt rejection of this modified membrane under high feed concentration was 99%, which was significantly higher than that of the unmodified one (70%). Additionally, in the absence of fouling, this modification increased water flux to 30 L/m^2h compared to the unmodified membrane with a flux of 12 L/m^2h.

The fourth ranked modification was coating the RO membrane with poly(methylacryloxyethyldimethyl benzyl ammonium chloride-r-acrylamide-r-2-hydroxyethyl methacrylate) [p(MDBAC-r-Am-r-HEMA)]. The modified membrane also achieved high salt rejection of 99% at high feed salt concentration. In the absence of fouling, the modified membrane flux was lower than that of the unmodified membrane only by 1 L/m^2h. However, the flux difference after fouling occurred indicated this modification had less fouling resistivity compared to other modified membrane.

Grafting with anionic 2-carboxylethyl acrylate ranked 5^{th} with a Pi value of 0.28. There was a significant gap in Pi (0.54) between rank 4 and 5. This was mainly due to testing this modified membrane with significantly lower feed salt concentration (7650 ppm) compared to the top 4 modifications. The flux difference after the fouling occurred was one of the highest among all 72 studies, 2.6 times higher than unmodified membrane.

Compared to the 72 modifications analyzed, the top four modifications exhibited a high performance despite being tested under high feed salt concentrations (> 30,000 ppm). In addition, only the top 4 modifications had Pi values greater than 0.80 compared to the majority of the studies with Pi values below 0.34 (Figure 2.9). These findings indicate that the top 4 modifications are promising for membrane fouling reduction. However, more testing and evaluation criteria (e.g., cost and scalability of the modification techniques) need to be considered to verify these findings.

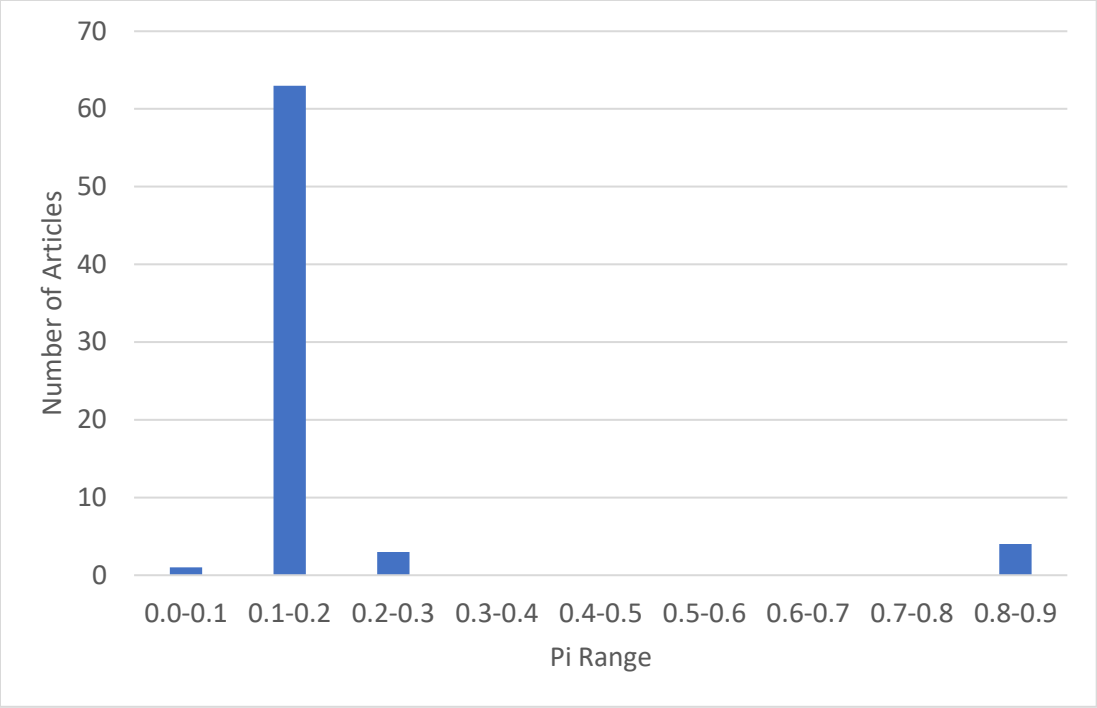


Figure 2. 9 Frequency distribution of the Pi values for the fouling studies.

3.2.2 Removal of NDMA

To rank the modifications for reducing NDMA concentration in the permeate, NDMA rejection was the evaluation criterion of highest importance followed by operation pressure and the improvement approach. Pressure was considered because it is one of the main reasons for the high operation and maintenance cost of RO systems. Since cost was never reported in any of the studies analyzed herein, pressure demand was used as an indicator for cost in the ranking process. Table 2.8 presents the weights of the evaluation criteria obtained from the AHP calculations. The consistency ratio was drastically lower than 0.1, confirming the validity of the AHP weighting results.

Table 2. 8 Weights of the evaluation criteria for NDMA rejection

Criteria	The AHP Weight
Rejection of NDMA	50%
Pressure demand	33%
Improvement Approach	17%

The criteria weights listed in Table 2.8 were used for calculating the Pi values using the TOPSIS method. The top 5 modifications for removal of NDMA are presented in Table 2.9. The modification of the operation conditions for the RO membranes ranked first. This study tested NDMA rejection for three membrane types under different condition for temperatures (15 – 30 °C) and pH (6 – 8). The NDMA rejection obtained by optimizing the operation conditions was 80% at 20 °C and pH of 8, which is a significant improvement compared to typical RO processes that has much lower NDMA rejection of 10 – 40% [57, 58]. However, this rejection level was not the highest among the top 5 modifications. Nonetheless, this modification approach ranked highest because it had the lowest pressure demand (145 psi) while achieving a significantly higher NDMA removal compared to typical RO processes. It is noted that 80% NDMA rejection may still not be sufficient to meet the safe levels of NMDA in water depending on the feed NDMA concentration. Therefore, future evaluations must consider adding the feed NDMA concentration to the evaluation criteria and including a threshold for acceptable NDMA removal.

Table 2. 9 Top 5 RO modifications for NDMA removal

Rank	Modification Type	Pi
1	Optimizing operation condition for NF90, SWC5, and low-pressure RO (TFC-HR) membranes	0.82
2	Heat-Treating ESPAB and HYDRApro501 Membranes	0.78
3	Optimizing feed temperature for RO and UF membranes	0.74
4	Low pressure RO membrane system modification	0.67
5	Full-scale RO filtration system pressure modification	0.60

Heat-treating HYDRApro501 and ESPAB membranes had the second highest Pi (0.78). This modification achieved NDMA removal of 98% while operating at a pressure of 223 psi. Optimizing commercial RO membrane performances under varying feed temperature for improving NDMA removal ranked 3rd with a Pi of 0.73. This approach resulted in NDMA removal of 70% at a pressure of 145 and 10 °C. Low pressure RO membrane system modification ranked 4th with a Pi of 0.67. This study achieved 60% NDMA removal using commercial RO membranes (ESPA2 and TFC-HR) operated at a pressure of 147 psi [157]. Full-scale RO filtration system with pressure modification ranked 5th and achieved 49% NDMA rejection at a pressure of 147 psi. Figure 2.10 presents the frequency distribution of all the NDMA removal studies analyzed. The study with the lowest Pi (0.17) achieved an NDMA rejection of 58% which was not the lowest among the studies. However, the pressure demand was 588 psi, which was the highest among the studies evaluated.

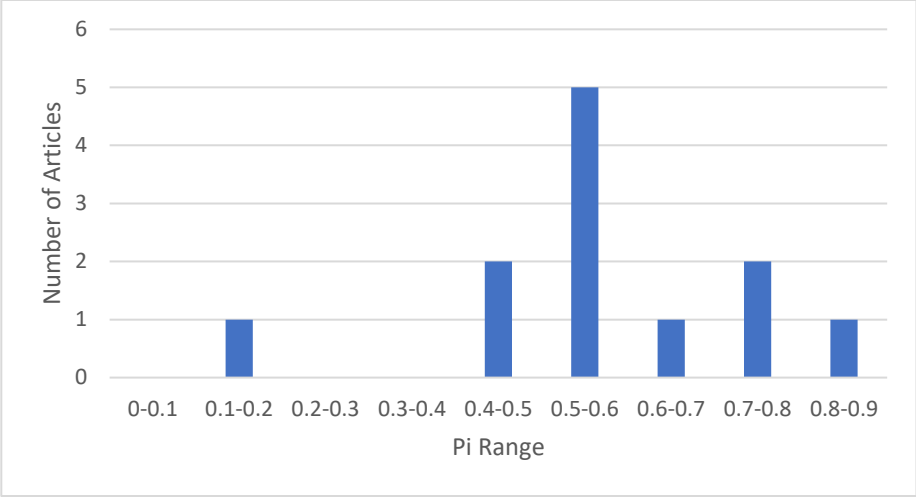


Figure 2. 10 Frequency distribution of the Pi for the NDMA removal studies

2.3.3 Sensitivity Analysis for Fouling Reduction

Assumptions were made to fill gaps for unreported data and to determine the weights for the evaluation criteria used in the MCDA process. Therefore, the rankings obtained for the membrane medications might have been biased because of the various judgments made to conduct this evaluation. A sensitivity analysis was performed to study the impact of the criteria weights on the ranking of the top 5 modifications identified previously.

2.3.3.1 Fouling Reduction

The feed salt concentration weight was increased from 11.1% to 31.1% in increments of 5% (Figure 2.11). The weights for the other evaluation criteria were decreased accordingly to maintain a total criteria weight of 100%. The order of the original top 5 modifications changed slightly; however, these modifications remained on the top 5 list for all weights tested (Table

2.10). This analysis indicates that the ranking obtained originally was not drastically sensitive to the feed salt concentration weighting

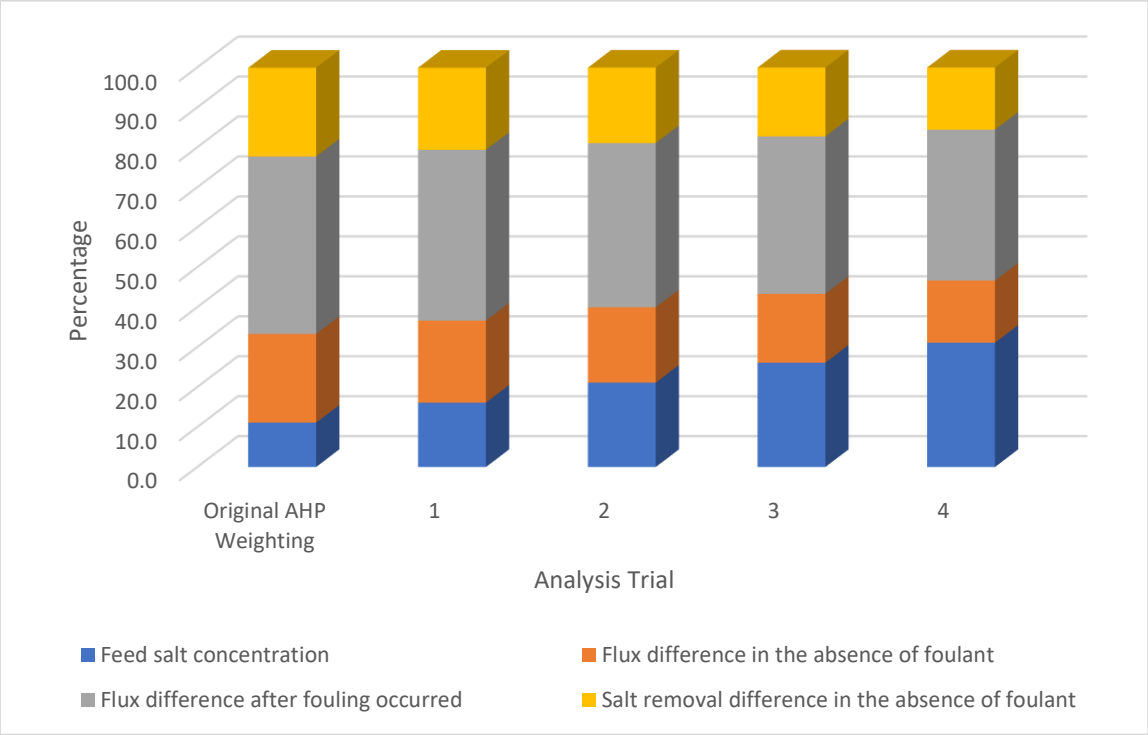


Figure 2. 11 Sensitivity test on weightings for the feed salt concentration criterion.

Table 2. 10 Sensitivity analysis results using different weightings for the feed salt concentration

Original Rank (11.1%)	**Pi**	**Trial 1 Rank (16.1%)**	**Pi**	**Trial 2 Rank (21.1%)**	**Pi**	**Trial 3 Rank (26.1%)**	**Pi**	**Trial 4 Rank (31.1%)**	**Pi**
1	0.88	3	0.77	3	0.69	3	0.64	3	0.61
2	0.87	4	0.75	4	0.68	4	0.63	4	0.61
3	0.83	1	0.82	1	0.72	1	0.66	1	0.63
4	0.81	2	0.81	2	0.71	2	0.66	2	0.63
5	0.22	5	0.30	5	0.39	5	0.48	5	0.57

The weight for the flux difference in the absence of foulant was increased from 22.2% to 42.2% (Figure 2.12). The weights for the other evaluation criteria were decreased accordingly to maintain a total criteria weight of 100%. Slight variation in order of the original top 5 modifications was observed but all of these modification remained on the top 5 list (Table 2.11). These results suggest that the assumptions made to obtain a weight for the flux difference in the absence of foulant did not considerably alter the overall MCDA results.

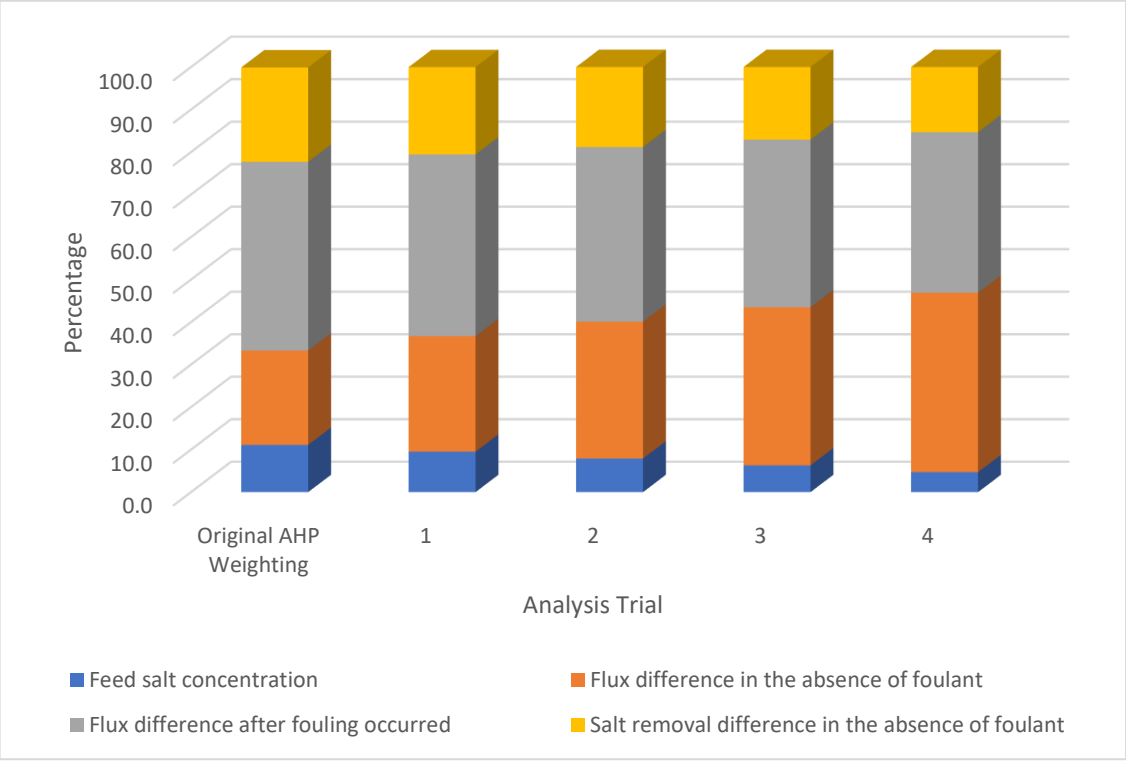


Figure 2. 12 Sensitivity test on weightings for the flux difference in the absence of foulant criterion.

Table 2. 11 Sensitivity analysis results using different weightings for the flux difference in the absence of foulant

Original Rank (22.2%)	Pi	Trial 1 Rank (27.2%)	Pi	Trial 2 Rank (32.2%)	Pi	Trial 3 Rank (37.2%)	Pi	Trial 4 Rank (42.2%)	Pi
1	0.88	2	0.79	2	0.67	2	0.54	2	0.41
2	0.87	1	0.81	1	0.69	1	0.56	1	0.42
3	0.83	5	0.71	4	0.60	4	0.48	4	0.36
4	0.81	3	0.73	3	0.61	3	0.49	3	0.37
5	0.27	4	0.19	5	0.16	5	0.13	5	0.11

The weight for the flux difference after fouling criterion was increased from 44.4% to 64.4% (Figure 2.13). The original top 5 modifications generally remained on this list (Table 2.12). However, the study originally ranked 5 dropped to rank 6 when the weight for the flux difference after fouling criterion was 64.4%. Overall, the original ranking for the membrane modifications was not highly sensitive to the variation in weight for the flux difference after fouling criterion.

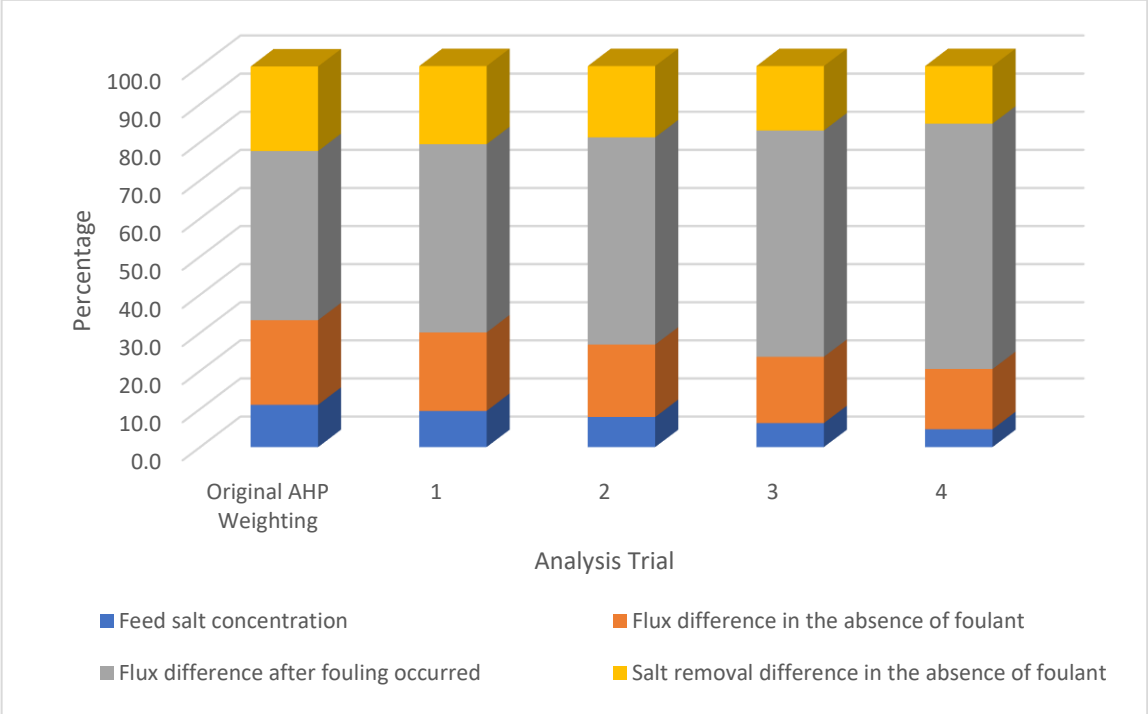


Figure 2. 13 Sensitivity test on weightings for the flux difference after fouling occurred criterion.

Table 2. 12 Sensitivity analysis results using different weightings for the flux difference after fouling occurred.

Original Rank (44.4%)	Pi	Trial 1 Rank (49.4%)	Pi	Trial 2 Rank (54.4%)	Pi	Trial 3 Rank (59.4%)	Pi	Trial 4 Rank (64.4%)	Pi
1	0.88	2	0.79	2	0.67	2	0.54	2	0.40
2	0.87	1	0.81	1	0.69	1	0.56	1	0.42
3	0.83	3	0.06	3	0.06	3	0.06	3	0.06
4	0.81	4	0.05	4	0.04	5	0.04	6	0.03
5	0.22	5	0.03	5	0.04	4	0.04	4	0.05

The weight for the salt removal difference in the absence of foulant criterion was increased from 22.2% to 42.2% (Figure 2.14). The original top 5 modifications remained on this list regardless of the weight evaluated (Table 2.13). However, the order of the top 5 modifications changed within this list.

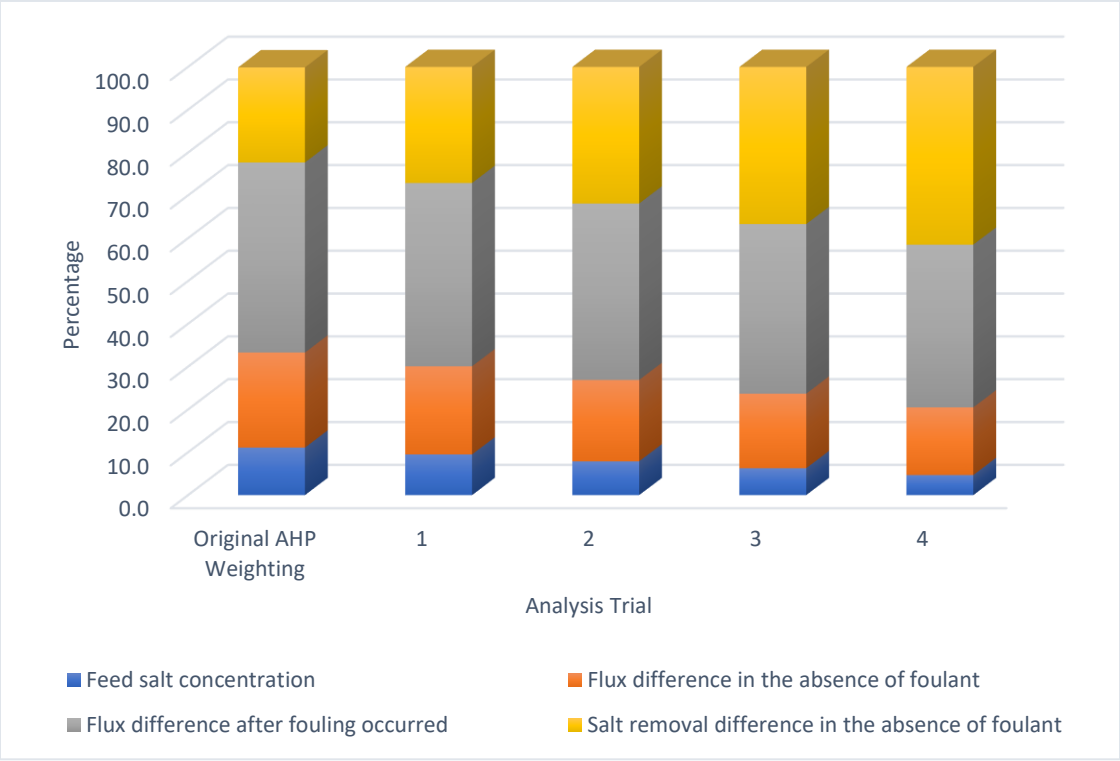


Figure 2. 14 Sensitivity test on weightings for the salt removal difference in the absence of foulant.

Table 2. 13 Sensitivity analysis results using different weightings for the salt removal difference in the absence of foulant.

Original Rank (22.2%)	**Pi**	**Trial 1 Rank (27.2%)**	**Pi**	**Trial 2 Rank (32.2%)**	**Pi**	**Trial 3 Rank (37.2%)**	**Pi**	**Trial 4 Rank (42.2%)**	**Pi**
1	0.88	2	0.79	2	0.67	2	0.54	2	0.40
2	0.87	1	0.81	1	0.69	1	0.55	1	0.42
3	0.83	4	0.71	4	0.59	4	0.48	4	0.36
4	0.81	3	0.73	3	0.61	3	0.49	3	0.37
5	0.22	5	0.19	5	0.16	5	0.14	5	0.11

A sensitivity analysis was also conducted to assess the impact of the assumptions made for assigning values for unreported data on the ranking of the top 5 modifications. In the original MCDA, all unreported values for a certain criterion were substituted with an "average value" that was determined based on the data reported in the other studies. In this sensitivity analysis, a "high value" and a "low value" were used to substitute unreported data. As shown in Table 2.14, the order of the original top 5 modifications changed but they remained on the top 5 list.

Table 2. 14 Sensitivity analysis for substitution of unreported data

Original Rank	**Pi**	**Rank based on "High Value"**	**Pi**	**Rank based on "Low Value"**	**Pi**
1	0.88	3	0.88	2	0.85
2	0.87	4	0.87	4	0.82
3	0.81	2	0.90	1	0.85
4	0.83	1	0.94	3	0.84
5	0.22	5	0.25	5	0.27

2.4 Conclusions and Future Recommendations

The majority of research on RO membrane modifications have been primality focused on fouling reduction. Surface coating and grafting with polymeric materials were the most utilized modification methods for fouling reduction. The research on RO modifications to remove of disinfection by products and their precursors is very limited despite the low rejection levels achieved by RO for the precursors and DBPs. The modifications alternatives were evaluated using the Analytical Hierarchy Process (AHP) and the Technique for Order Preference by Similarity to Ideal Solution (TOPSIS) to rank their performance based on a number of criteria. The top 5 ranked modification alternatives for fouling reduction were identified. These modifications utilize coating and grafting techniques to alter the surface hydrophobicity and pore size of the RO membrane to make it less prone to fouling. For improving NDMA removal, modifications beyond altering the membrane surface properties were used. For example, modifying the operation conditions of the RO system (e.g., pressure) of the RO system configuration (e.g., multiple stage RO) resulted in drastically higher NDMA removal. However, half of the research on modifications reported NDMA rejection of 80% or below. Although these rejection levels are drastically higher than the 10 – 40 % rejection achieved traditionally by RO membranes, more research is needed to improve NDMA rejection to sufficient levels to ensure safe production.

The MCDA conducted herein highlighted the most promising modifications to reduce fouling and improve NDMA rejection. Further research is needed to overcome the limitations of such promising approaches to make them viable alternatives for large-scale use. A sensitivity analysis

was conducted to realize the impact of varying the weights of the evaluation criteria on the ranks of the modifications. Overall, the top 5 modifications remained on this list. However, the outcomes of the MCDA should be further refined and verified in future research by including more evaluation criteria to conduct the analysis. This was not possible in this research because the studies evaluated have limited data on important evaluation criteria such as cost, toxicity, and sustainability considerations.

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Appendix

Table A. 1 TOPSIS analysis of the data of fouling reduction

11.10%	22.20%	44.40%	22.20%	Si +	Si -	Pi	Rank
Water Characteristic	Flux Reduction Before Fouling	Flux Reduction Resistivity	Salt Rejection				
0.003	0.002	0.003	0.003	0.055	0.003	0.056	
0.003	0.002	0.005	0.003	0.055	0.011	0.170	
0.003	0.003	0.004	0.003	0.055	0.011	0.166	
0.058	0.001	0.003	0.003	0.008	0.059	0.876	2
0.003	0.002	0.003	0.003	0.055	0.010	0.159	
0.003	0.001	0.007	0.003	0.055	0.012	0.185	
0.001	0.003	0.004	0.003	0.058	0.011	0.156	
0.003	0.005	0.004	0.003	0.055	0.012	0.177	
0.003	0.004	0.004	0.003	0.055	0.012	0.175	
0.003	0.003	0.003	0.003	0.055	0.010	0.157	
0.002	0.005	0.004	0.003	0.057	0.012	0.172	
0.002	0.002	0.005	0.003	0.056	0.011	0.167	
0.003	0.003	0.003	0.003	0.055	0.011	0.165	

11.10%	22.20%	44.40%	22.20%				
Water Characteristic	**Flux Reduction Before Fouling**	**Flux Reduction Resistivity**	**Salt Rejection**	**Si +**	**Si -**	**Pi**	**Rank**
0.003	0.001	0.005	0.003	0.055	0.011	0.162	
0.056	0.003	0.003	0.003	0.008	0.057	0.883	1
0.004	0.003	0.004	0.003	0.054	0.011	0.170	
0.003	0.003	0.006	0.003	0.055	0.013	0.192	10
0.003	0.003	0.003	0.003	0.055	0.011	0.161	
0.001	0.002	0.004	0.003	0.058	0.011	0.156	
0.003	0.003	0.004	0.003	0.055	0.011	0.169	
0.051	0.004	0.003	0.003	0.010	0.052	0.835	3
0.012	0.002	0.007	0.003	0.046	0.018	0.275	5
0.001	0.003	0.003	0.003	0.058	0.010	0.153	
0.003	0.003	0.004	0.003	0.055	0.011	0.164	
0.003	0.004	0.003	0.003	0.055	0.011	0.167	
0.000	0.003	0.004	0.003	0.058	0.011	0.154	
0.003	0.002	0.004	0.003	0.055	0.011	0.166	
0.003	0.003	0.003	0.003	0.055	0.011	0.162	
0.004	0.003	0.004	0.004	0.055	0.012	0.175	
0.001	0.003	0.004	0.003	0.057	0.011	0.160	

11.10%	22.20%	44.40%	22.20%	Si +	Si -	Pi	Rank
Water Characteristic	**Flux Reduction Before Fouling**	**Flux Reduction Resistivity**	**Salt Rejection**				
0.002	0.004	0.003	0.003	0.057	0.011	0.163	
0.003	0.002	0.004	0.003	0.055	0.011	0.169	
0.003	0.003	0.003	0.003	0.055	0.011	0.162	
0.003	0.002	0.003	0.003	0.055	0.010	0.155	
0.003	0.003	0.003	0.003	0.055	0.011	0.168	
0.003	0.001	0.003	0.003	0.055	0.010	0.148	
0.008	0.001	0.003	0.003	0.051	0.012	0.191	
0.050	0.003	0.003	0.003	0.011	0.051	0.815	4
0.000	0.003	0.003	0.003	0.058	0.010	0.151	
0.001	0.003	0.004	0.003	0.058	0.011	0.157	
0.003	0.002	0.003	0.003	0.055	0.010	0.154	
0.003	0.003	0.004	0.003	0.055	0.011	0.167	
0.003	0.002	0.005	0.003	0.055	0.012	0.173	
0.003	0.001	0.003	0.003	0.055	0.010	0.148	
0.004	0.002	0.003	0.003	0.054	0.011	0.165	
0.004	0.003	0.003	0.003	0.054	0.011	0.171	
0.003	0.002	0.005	0.003	0.055	0.011	0.167	
0.003	0.003	0.003	0.003	0.055	0.010	0.159	
0.003	0.003	0.006	0.003	0.055	0.012	0.184	
0.001	0.003	0.006	0.003	0.058	0.012	0.168	
0.002	0.003	0.006	0.003	0.057	0.012	0.176	
0.001	0.002	0.010	0.003	0.057	0.015	0.211	7
0.003	0.004	0.003	0.003	0.055	0.011	0.168	
0.003	0.003	0.003	0.003	0.055	0.011	0.162	
0.003	0.003	0.008	0.003	0.055	0.014	0.200	9

11.10%	22.20%	44.40%	22.20%				
Water Characteristic	**Flux Reduction Before Fouling**	**Flux Reduction Resistivity**	**Salt Rejection**	**Si +**	**Si -**	**Pi**	**Rank**
0.003	0.004	0.001	0.003	0.056	0.010	0.154	
0.004	0.001	0.003	0.003	0.054	0.010	0.158	
0.003	0.003	0.004	0.003	0.055	0.011	0.165	
0.001	0.001	0.003	0.003	0.058	0.009	0.137	
0.003	0.003	0.003	0.003	0.055	0.011	0.163	
0.003	0.003	0.006	0.003	0.055	0.013	0.186	
0.003	0.002	0.010	0.003	0.055	0.015	0.219	6
0.003	0.001	0.003	0.003	0.055	0.010	0.148	
0.009	0.002	0.003	0.003	0.050	0.013	0.213	8
0.003	0.001	0.000	0.003	0.056	0.008	0.130	
0.003	0.005	0.005	0.003	0.055	0.013	0.188	
0.003	0.005	0.003	0.003	0.055	0.012	0.176	
0.003	0.002	0.005	0.003	0.055	0.011	0.171	
0.003	0.003	0.001	0.004	0.056	0.010	0.151	
0.003	0.005	0.003	0.003	0.055	0.012	0.178	
0.003	0.001	0.004	0.003	0.055	0.010	0.155	

Table A.2 TOPSIS analysis of the articles on NDMA removal

Removal	Pressure	Improvement	Si+	Si-	Pi	Rank
0.19	0.17	0.00	0.15	0.10	0.41	
0.18	0.16	0.00	0.14	0.10	0.41	
0.08	0.04	0.04	0.12	0.13	0.51	
0.12	0.17	0.00	0.17	0.04	0.17	
0.08	0.04	0.04	0.12	0.13	0.52	
0.10	0.04	0.08	0.10	0.15	0.60	5
0.11	0.06	0.04	0.10	0.11	0.54	
0.17	0.11	0.04	0.09	0.11	0.55	
0.20	0.06	0.04	0.05	0.16	0.78	2
0.14	0.04	0.04	0.05	0.15	0.73	3
0.16	0.04	0.08	0.04	0.17	0.82	1
0.12	0.04	0.08	0.08	0.15	0.67	4